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QUANTUM OPTICS

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QUANTUM OPTICS

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Directors' Note

The 1969 Scottish Universities Summer School in Physics was the tenth, in what is now a well-established annual series. The formal lecture programme is fully presented in this volume, and in addition we have included a selection from the seminars in which participants in the school—students and staff—spoke about their own work and interests. Three visitors to the school—Professors A. Kastler, L. Mandel and A. L. Schawlow—also gave seminars which were enthusiastically appreciated, but it has not been practicable to include them here.

It was a particular source of pleasure to all of us at this 10th School that Professor Kastler, accompanied by Madame Kastler, spent ten days with us, sharing in all our activities, academic, recreative and social.

The devoted work of the editors, Susan Kay and Arthur Maitland, is attested by this volume. We are glad to be able to acknowledge here the great contribution made to the School by the Secretary, Dr. A. G. A. Rae, the Treasurer, Dr. A. L. S. Smith, and the other members of our Committee.

P. S. Farago
R. M. Sillitto } Joint Directors.

Editors' Note

Each of the chapters in this book covers the material delivered by the lecturers at the Summer School and has been largely prepared by them with the minimum of editing. The chapter by Professor Glauber has been prepared by Mr. A. Maitland from tape recordings together with notes taken by Dr. D. F. Walls and Mr. S. S. Rangnekar. Professor Glauber kindly gave his permission for a verbatim transcript to be taken in an attempt to bring the tutorial atmosphere and spirit of a summer school to the more usual cold print of published proceedings. The lectures given by Professor Louisell closely followed those given at the 1967 Enrico Fermi Summer School, Course 42. Dr. Pike's chapter is based substantially on a review presented at the inaugural meeting of the European Physical Society. The articles by Haken *et al.*, Graham, Walls, Haake, Peřina and Bénard were presented as seminars. Some of these have been revised by the Editors.

The following lectures were also given at the School, but have not been included in the Proceedings. Professor A. Kastler delivered two lectures on "Optical Pumping". Professor A. L. Schawlow gave a seminar on "The Laser—Present and Future", and Professor L. Mandel presented a paper on "The Interference of Photon Beams at Low Light Intensities".

Whilst most of the authors have seen their manuscripts in proof, the Editors accept responsibility for any errors and misprints.

A.M.
August 1970 S.M.K.

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Introductory Remarks

R. M. SILLITTO

Quantum optics began when quantum physics began—or perhaps slightly before, since quantum optics initiated quantum physics. The investigation of the black-body spectral distribution, and its interpretation with the help of the statistical mechanics of radiation, led to the hypothesis of quantization, while Einstein's interpretation of the photoelectric effect introduced the notion of the photon. The correlation of the statistical and spectral characteristics of radiation recurs again and again as one of the central tasks of quantum optics, and the statistics of photo-emission are primary data for the discussion of the statistical and coherence properties of light-fields.

But quantum optics, as we know it, hardly existed until the 1950s—though the lack of it had struck some people as odd. Glauber summed this up at Rochester in 1966 when he remarked that quantum theory owed a lot to optics, but had only recently begun to repay the debt. At the beginning of this volume on Quantum Optics, it is worth while giving a little space to a consideration of this curious historical phenomenon.

Physical optics has, in many ways, been remarkably unperturbed by the two major achievements of 20th century physics. Maxwell's equations, extrapolated into a frequency domain far removed from that which provides their experimental basis, are the foundation of theoretical physical optics. For very profound reasons concerned with the intimate relationship between the electric and the magnetic field, these equations were already Lorentz invariant, and so survived the advent of special relativity. And the extrapolation to frequencies 10^{10} or more times those of Faraday's experiments on electromagnetic induction proved adequate in all respects save one. That one deficiency was certainly important—it needed the hypothesis of quantization to remedy it. But this necessity appears in optics only when we are concerned to discuss the processes of emission and absorption, and such related processes as dispersion. By relegating the details of these discussions to atomic physics, physical optics remained serenely above the battle.

In large part, the reliance which those who developed quantum theory could place on optical evidence stemmed from the apparent inviolability of physical optics. Attempts to find quantum effects in the physical-optical fields of diffraction and interference—by Taylor[1] in 1909, by Dempster and Batho[2] in 1927, and by Jánossy and Náray[3] in 1957—were convincingly

negative,† and served two distinct and important purposes: firstly, they confirmed the reliability at all intensity levels of the interferometric evidence on which optical spectroscopy depends; and secondly, they provided the experimental basis for the interpretation of the wave-function as giving probabilities relating to a single particle, rather than describing directly the probability-density of particles. It is worth remarking here that interference experiments prior to 1960 were always carried out at "low intensities", in the sense that the degeneracy parameter of the radiation employed—that is, the number of quanta per coherence volume—was always less than unity by two or more orders of magnitude. Lacking this critical measure of intensity the experimenters mentioned above used other criteria for deciding how low the intensity had to be for their experiments to be significant—for instance, that the average energy within their interferometers at any time should be less than a quantum. In retrospect this requirement seems unnecessarily severe, since the crucial question is whether the degeneracy parameter is, or is not, much less than unity.

Quantum effects in optical processes were sought in other ways, and Campbell[5], with rare perception, attempted an intensity-correlation experiment in 1909. The experiment was reported in two fascinating papers, the first of which began: "This is an account of an experiment which failed..."; and the second reported that, "The experiment has continued unsuccessful." Campbell was not the last person to be unsuccessful with an intensity-correlation experiment, although Bay and Farago[6] have pointed out that some of the photon-coincidence experiments which produced null-results were attempts to detect an effect linear in the intensity: such an effect, if it existed, would not be an intensity correlation.

As long as optical detectors were long-time-constant devices—and "long-time" here means so long, that many quanta are detected within the response time—quantization does not need to be considered, though the broader statistical characteristics of the radiation do. So coherence theory was developed to take into account the statistical and spectral features of the light emitted by the high-frequency noise-generators which optical experimenters used as light sources. The random process of a light-beam was assumed to be Gaussian. The justification for this assumption was not complete, but once the assumption was made it allowed attention to be concentrated on the second moment of the probability distribution of the light-field. The measurements of what was called the second-order coherence function (and is sometimes now called the *first*-order coherence function) determined the single parameter of the Gaussian distribution. In the years after the war, Wolf,

† A reported positive result by Dontsov and Baz (1967)[4] cannot yet be regarded as established.

Hopkins and others developed the classical coherence theory—which had originated as early as 1855[7]—into a theory of physical optics in terms of observables[8]; the unobservable time-variations which underlie elementary wave-optical theory had been eliminated. The resemblance between coherence theory in this form, and the theory of random signals in radio engineering and communications theory, now became clear and fruitful. This merging of languages and modes of thought has had important consequences in radio and optical astronomy, in studies of optical imaging, and in the developing technology of information storage and retrieval.

The development of coherence theory to the point where it became a classical theory of optics in terms of observables has, in retrospect, a two-fold significance for quantum optics. Firstly, it prepared the ground for the introduction of a quantum theory, which has been developed since 1963 in the work of Glauber and others; and secondly, it established a great domain of optical theory which must survive any quantum reappraisal.

In a sense, this latter point seemed to have been anticipated in a very general way by Heisenberg in 1930, when he established "that the classical wave theory is sufficient for the discussion of all questions of coherence and interference". But this dictum seems to have been forgotten—or at least overlooked —when the experiments of Forrester, Gudmundsen and Johnson, and of Hanbury-Brown and Twiss, aroused controversy and challenged explanation. In classical terms, the results of both these experiments were easily explained—except, of course, that the use of photoelectric detectors was vital to both experiments, and the explanation of photoelectric emission must be quantal. Both experiments exhibited the intensity correlation phenomenon Campbell had sought almost fifty years before—in the one case correlations in time, and in the other case correlations in space. The argument that this violated Dirac's dictum, that a photon "interferes only with itself", arose from a too naive idea of what is implied by the term "photon", and has been very effectively answered by, amongst others, Hanbury-Brown and Twiss, and Mandel. What emerges very clearly from a reading of the papers written while the debate was still keen, is the necessity—in an era of photomultiplier tubes and fast electronic circuitry—for a theory which allows the discussion of optical coherence and of the interaction of matter and radiation to be carried on within a single formalism. The separation of physical optics and of atomic physics could no longer be maintained, and a quantum theory of optical coherence was urgently needed.

Meanwhile other strands of the fabric of quantum optics were being spun. Microwave engineering, statistical mechanics, and the quantum theory of matter came together in the invention of the maser in 1954, and quantum electronics began to emerge as a recognizable discipline. The negative resistance characteristic, which is necessary for the possibility of oscillation, is

achieved by the production of a population inversion in the active medium, and quantum electronics has been defined as the study and exploitation of population inversion. The induced and spontaneous emissions from the upper maser level provide the signal and the inherent noise respectively in the maser, and the term "quantum noise" was coined to refer to spontaneous emission in this context. The importance of the maser was at once seen to be its extremely good signal: noise capability. Simple considerations on the transition probabilities for spontaneous and induced emission as functions of frequency showed that the inherent signal:noise ratio of maser devices at optical frequencies might be much less favourable than in the microwave region, and it was six years before the successful operation of the laser laid the doubts to rest.

The successful operation of the laser in 1960 brought a new series of problems; one set, concerned with the nature of laser light, and the features which distinguish it from "ordinary" or thermal light, the other set, concerned with the way the laser itself works. The two sets of problems are obviously related, since, on the one hand, the peculiar features of the output light are consequences of the way the device works, and, on the other hand, experimental investigations of the statistics of the photoelectron emission produced by laser light furnish data for testing theories of the mechanism of the laser. It turns out that a classical model derived from the Van der Pol treatment of the triode oscillator provides a useful phenomenological theory of the laser as a non-linear oscillator driven by noise—the spontaneous emission from the upper laser level. Both intensity-noise and phase-noise are predicted by this model, in terms of a very small number of model parameters, and the model provides a useful basis for the discussion of experimental results on the photon statistics of laser beams. The detailed description of the working of the laser, in terms of the processes operating at the microscopic level, is a difficult exercise in the quantum mechanical non-equilibrium statistical thermodynamics of an open system.It has been approached in stages—firstly, using quantum mechanics to describe the atoms in the laser-active medium, and classical electromagnetic theory to describe the radiation field; then, by adding to this description a random polarization to describe, phenomenologically, the effects of spontaneous emission; and finally, using the quantized field theory and the Fokker–Planck equation to describe the build-up of the radiation field. This work is able to account for the observed intensity fluctuations in lasers operating both below and above threshold, and for the observed variation of bandwidth with pump power; in particular, the relative contributions of phase-noise and intensity-noise can be understood. A very different treatment in terms of rate-equations accounts satisfactorily for the observations on intensity fluctuations, but does not include the effects of phase-diffusion.

The role of the phenomenon of "super-radiance" in laser operation has recently attracted attention. The extension of the Weisskopf–Wigner theory of

natural line-width, to systems of many atoms in the same excited state, predicts emission phenomena which, depending on the density and configuration of the excited ensemble, may display some of the features characteristic of laser emission, or may in other circumstances exhibit the characteristics of the radiation-trapping which is observed in resonance-fluorescence. In the one, super-radiant, case the radiative lifetime of the ensemble is much less than that of a single excited atom, while in the other, sub-radiant, case the lifetime may be greatly increased. The terms "induced emission" and "spontaneous emission" do not seem to be appropriate any longer in discussing the radiative properties of such ensembles. The relationship between this approach to laser theory and the approach using the Fokker–Planck equation raises a number of interesting and fundamental questions.

The Fokker–Planck equation treatment, which has now been very fully worked out, leads to detailed predictions about the build-up in time of the laser-field, the intensity fluctuations and line-width at different power levels, and—as a corollary of this—the coherence properties of the emitted light. Haken has pointed out that the laser light-field "appears as a carrier of the quantum fluctuations of the atoms, which (fluctuations) are filtered and transformed by a non-linear process". The study of these intensity fluctuations, or, equivalently, of the photon-statistics of the light-field, is thus a test of the microscopic theory and, besides, gives an insight into the nature of the light-field. To describe the properties of such fields, the theory of coherence, as it evolved in relation to the Gaussian light-fields of thermal light sources, has had to be extended and refined, additional statistical parameters being related to a hierarchy of coherence functions defined in slightly different ways by different authors.[9] The results of multidetector correlation experiments can be expressed in terms of these coherence functions and by a suitable ordering of creation and annihilation operators one can construct appropriate correlation functions to describe multiple coincidence rates for either photodetectors or stimulated -emission photon counters. As yet, little experimental work has been done on the higher-order correlations which are revealed either by triple- and higher-order coincidence experiments, or by photon-statistics experiments, but enough has been done to show that there is significant new information to be obtained in this way.[10] Meanwhile, intensity correlation experiments, in which photon-coincidences are observed or photon-statistical distributions are determined, have provided the means of measuring optical line-widths orders of magnitude narrower than could be measured by optical interferometry.

Even before the advent of the laser, very long Fabry–Perot etalons were constructed for spectrometry, and for use as very narrow-band optical filters. For instance, a 156 cm etalon was in use at NBS in the late 1950s for measurements of the spectral line-width of an atomic beam light source, and Connes

used an etalon 260 cm long as a filter to produce a very narrow spectral line for experiments on light-modulation, in about 1960. The use of Fabry–Perot systems as laser resonant cavities has sparked extremely interesting and fruitful investigations of the physical optics of the Fabry–Perot resonator, both when it functions passively—as interferometer or filter—and when it functions actively, with excited matter in the cavity. Computer solutions of the electromagnetic field equations in the cavity have revealed subtle complexities in its behaviour which were previously unsuspected, and both the parallel-plate system and a variety of confocal and non-confocal systems using mirrors or lenses have been studied; some of these systems have unfamiliar forms (see, e.g. Toraldo di Francia's Chapter in this volume). The mathematics of these studies is elegant in itself, and in practical terms the development of the confocal resonator system and its derivatives has a lot to do with the ruggedness and reliability of presently available gas lasers. The discussion of the properties of cavities containing active material with a complex dielectric constant is, of course, one of the possible avenues of approach to laser dynamics.

While the maser was being developed as a low-noise microwave amplifier, microwave engineers and physicists were showing a revived interest in the potentialities of an alternative device, the parametric amplifier. The principle of the energy-conversion mechanism of the parametric amplifier was illustrated by phenomena observed by Faraday,[11] and Melde,[12] and was analyzed and systematically experimented with by Lord Rayleigh.[13] It was used in a radio-telephone system between Berlin and Vienna before 1915, and Lee de Forest, the inventor of the triode, remarked in 1916 that, at that time, the "magnetic amplifier", as the then-current form of parametric amplifier was called, had advantages over the triode at high power levels. However, he predicted that the development of high-power triodes might soon change this situation, and in this he was right. Interest in the parametric amplifier then waned, although a number of communications theorists, among them R.V.L. Hartley, continued to study the principle, and in 1928 Hartley suggested, at a meeting of the American Physical Society, that the Raman effect could be understood in terms of the same general coupling and frequency-mixing mechanism as the parametric amplifier.[14] The rates of energy transfer in these systems are described by a set of very general relations, due to Manley and Rowe, which seem to apply wherever the coupling device or medium is a non-linear reactance.

The theory of quantum mechanical oscillators and attenuators, which originated in the 1950s, can be used to discuss such systems as an idealized maser, or a parametric amplifier, or frequency converter[15] and it has not always been clear where the dividing line between these devices should be drawn—or, indeed, if such a dividing line is necessary or useful. The ele-

mentary process involved in the optical parametric amplifier, oscillator, or converter, is sometimes referred to as a three-boson process, although the term should not be taken too literally. Parametric processes may be phase-dependent, and this means that the fields involved should be represented by coherent states—or, if the intensities are high enough, by classical waves—and these are states in which the numbers of quanta are ill-defined. However, the term is useful as far as it helps in the classification of the various phenomena which may appear; thus, the annihilation of a photon, accompanied by the creation of a photon of different frequency or polarization and a phonon, is characteristic of the stimulated Raman or Brillouin effect, while the annihilation of one photon and the creation of two photons is the elementary mechanism of the optical parametric oscillator or amplifier. The latter has the interesting and potentially useful property of being tunable over a quite considerable wavelength range—tuning ranges of up to 3:1 in wavelength, and output powers up to 1kW, have been reported.[16]

In these devices, the parameter which couples the oscillating fields of different frequencies is a field-dependent polarizability,[17] so the mechanisms involved lie within the domain of non-linear optics. The advent of the laser, with its very high field strengths, has made it possible to exhibit and study a great variety of very striking effects in solids, liquids, gases and plasmas. In solids, these effects provide the solid-state physicist with a powerful armoury for the study of elastic behaviour and its structure-dependence. In particular, the crystal structure changes at phase transitions modify the non-linear optical behaviour. The theory of non-linear effects in solids has been very highly developed to treat interactions with phonons, magnons, polaritons and other excitations. The behaviour of sound waves in liquids, critical point phenomena, and molecular excitation and vibration processes have been studied, along with various electrical and electromagnetic phenomena such as dielectric breakdown, and multiphoton excitation. Some of the optical effects are very beautiful in their own right—the ring patterns produced in induced Raman scattering, for instance, and the very striking phase-matching process which Giordmaine and Terhune discovered in negative uniaxial crystals. To the experimenter at least, the evident beauty of so many optical phenomena has always been one of the appealing aspects of optics, and these effects are surely in this tradition.

It is often the case that new tools and techniques are vigorously applied and exploited even while their structure and mode of operation are still being studied. This happens here: the laser is used to generate non-linear optical processes, and thus we learn to understand better the effect of the non-linear properties of the active material in the laser cavity itself.

In many respects, the elementary processes which are important in quantum optics are exhibited most directly in optical pumping experiments. Here, inter-

actions are produced between "free" atoms and one or more oscillatory fields, so that, for instance, one- or two-photon transitions can be studied without introducing the complexities of condensed matter. But even in tenuous vapours the "free" atom is a fiction—all the atoms present are coupled through their interaction with the radiation field, and this coupling provides a rich harvest of effects for study and elucidation.

The term "pompage optique" was introduced in 1950 by Kastler to describe a particular method of producing a population inversion, but there are affinities between modern optical pumping experiments and experiments on the magnetic depolarization of resonance radiation by Wood and Ellett[18] and by Hanle[19] in 1923, and the subsequent experiments on the effect of radio-frequency fields on the polarization of resonance radiation by Fermi and Rasetti[20] and by Breit and Ellett.[21] The Hanle experiment is a special case of a level-crossing experiment, exhibiting optical effects arising from the coherent superposition of excited quantum states. In the "coherent trapping" process this quantum mechanical—or "Hertzian"—coherence is transferred by the radiation field from one atom to another or others. It is not obvious that the radiative interaction should be the only one which can transfer Hertzian coherence between atoms, and there are various experiments which show that it can be transferred in collisions.[22]

There is an idication here of the very intimate connection between optical coherence and atomic properties. This connection obviously underlines the mechanism of coherent emission in the laser, and in optical parametric devices, but the optical pumping experiments seem to show it in its most elementary form. Considerable interest attaches to further studies of these effects, therefore, and to theoretical work such as that of Dicke,[23] Barrat[24] and Ernst and Stehle,[25] which relates them to the process of coherent emission for more complex ensembles.

The past fifteen or twenty years has seen a veritable renaissance in optics. This has happened before. For instance, in 1900 Drude wrote that in optics "there pulses a new life whose further nourishing must be inviting to every one". In 1956, Zernike said at a meeting in Boston that, activity in optics proceeds in waves, and that at that time, optics was emerging from a trough and heading for a peak. In large part the enormous wave of activity on whose crest we now seem to be riding has been stimulated by the merging of a rich variety of streams of thought and technique. It is the purpose of this volume to present articles which will clarify in the readers' minds the nature and interrelations of some of the major influences in this creatively non-linear process.

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Quantum Electrodynamics

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Quantum electrodynamics is the theory which describes the interaction of electrons (either free or bound in atoms) with the electromagnetic field, or equivalently with photons. In the complete theory, both the electrons and the field are treated quantum mechanically, but for some purposes a classical treatment of one or the other may suffice. In what is generally called semiclassical theory, the electrons are treated quantum mechanically, but the field is regarded as an externally prescribed classical field. However, in certain problems we may reverse this procedure and treat the field quantum mechanically, while replacing the electrons by a classical current distribution, externally prescribed.

We shall begin by reviewing briefly the Hamiltonian formalism in both classical and quantum mechanics, using the harmonic oscillator as an example. Then we shall go on to deal with semiclassical theory, applied particularly to induced emission and absorption of radiation by atoms. Next, we shall discuss the quantization of the electromagnetic field, and its interaction with a classical current distribution. Finally, we turn to the full theory of quantum electrodynamics.

1. HAMILTONIAN FORMALISM

1.1 CLASSICAL MECHANICS

The time development of a classical system whose instantaneous state is specified by the generalized coordinates and generalized momenta $q_1, \ldots, q_n$ and $p_1, \ldots, p_n$ is described by Hamilton's equations

$$\frac{\mathrm{d}q_j}{\mathrm{d}t} = \frac{\partial H}{\partial p_j}, \quad \frac{\mathrm{d}p_j}{\mathrm{d}t} = -\frac{\partial H}{\partial q_j}, \quad (j = 1, \ldots, n) \tag{1}$$

where H is the Hamiltonian function, a function of the coordinates and momenta, and possibly also explicitly of the time. Since the coordinates and momenta are themselves functions of time, the rate of change of H is

$$\frac{\mathrm{d}H}{\mathrm{d}t} = \frac{\partial H}{\partial t} + \sum_{j=1}^{n} \left\{ \frac{\partial H}{\partial q_j} \frac{\mathrm{d}q_j}{\mathrm{d}t} + \frac{\partial H}{\partial p_j} \frac{\mathrm{d}p_j}{\mathrm{d}t} \right\} = \frac{\partial H}{\partial t}$$

since by (1) the terms in the sum cancel. If H is not explicitly time-dependent, then it is a constant of the motion, whose value is the total energy. The appearance of an explicit time-dependence means that the system is subjected to external forces.

For example, a forced harmonic oscillator (in one dimension) is described by the Hamiltonian function

$$H = \frac{1}{2m} p^2 + \tfrac{1}{2} m\omega^2 q^2 - q\,\mathrm{f}(t), \tag{3}$$

where $\mathrm{f}(t)$ is a given function of time. Hamilton's equations are

$$\frac{\mathrm{d}q}{\mathrm{d}t} = \frac{\partial H}{\partial p} = \frac{p}{m}, \qquad \frac{\mathrm{d}p}{\mathrm{d}t} = -\frac{\partial H}{\partial q} = -m\omega^2 q + \mathrm{f}(t). \tag{4}$$

It is convenient to introduce in place of q and p a complex variable

$$z = m\omega q + ip. \tag{5}$$

It satisfies the equation of motion

$$\mathrm{d}z/\mathrm{d}t = -i\omega z + i\,\mathrm{f}(t). \tag{6}$$

If there is no external force, then the motion corresponds to a circle, described with constant angular velocity $-\omega$, in the complex z plane (which may essentially be identified with the two-dimensional classical phase space). More generally, the solution of (6) is

$$z(t) = z_0 \exp(-i\omega t) + i \int_{-\infty}^{t} \mathrm{d}t'\, \mathrm{f}(t') \exp[-i\omega(t-t')], \tag{7}$$

where z_0 is an arbitrary complex constant. In particular, if f vanishes except during some finite time interval $t_0 < t < t_1$, then

$$z(t) = z_0 \exp(-i\omega t) \text{ for } t < t_0$$

and

$$z(t) = z_1 \exp(-i\omega t) \text{ for } t > t_1,$$

where the constants z_0 and z_1 are related by

$$z_1 - z_0 = i \int_{t_0}^{t_1} \mathrm{d}t\, \mathrm{f}(t) \exp(i\omega t) = i\,\tilde{\mathrm{f}}(\omega). \tag{8}$$

When $\mathrm{f} = 0$, H is constant, and equal to $|z|^2/2m$.

1.2 REVIEW OF QUANTUM MECHANICS

In quantum mechanics, the coordinates and momenta are replaced by operators $\hat{q}_1, \ldots, \hat{q}_n$ and $\hat{p}_1, \ldots, \hat{p}_n$ which satisfy the commutation relations

$$[\hat{q}_i, \hat{q}_j] = [\hat{p}_i, \hat{p}_j] = 0, \qquad [\hat{q}_i, \hat{p}_j] = i\hbar\delta_{ij}, \tag{9}$$

where $[\hat{A}, \hat{B}] = \hat{A}\hat{B} - \hat{B}\hat{A}$, and where δ_{ij} is the Krönecker delta symbol defined by

$$\delta_{ij} = \begin{cases} 1 \text{ if } i = j, \\ 0 \text{ if } i \neq j. \end{cases}$$

In one particular representation (the q representation), these operators act on functions $\psi(q_1, \ldots, q_n)$, according to the relations

$$\hat{q}_i\psi = q_i\psi, \qquad \hat{p}_i\psi = -i\hbar\frac{\partial}{\partial q_i}\psi. \tag{10}$$

We prefer however not to choose a particular representation, but to use the more abstract Dirac notation, in which the states are represented by state-vectors $|\psi\rangle$ on which the operators act. There is defined a scalar product $\langle\phi|\psi\rangle$ between any two such state-vectors, satisfying $\langle\phi|\psi\rangle^* = \langle\psi|\phi\rangle$, where the star denotes complex conjugation. (In the q representation, the scalar product is the integral over all q_i of $\phi^*\psi$.) Two state-vectors $|\phi\rangle$ and $c|\phi\rangle$ which differ only by an overall multiplicative factor describe the same state. It is possible to normalize the state-vectors so that $\langle\phi|\phi\rangle = 1$, but even then there remains an arbitrary phase factor in $|\phi\rangle$.

For any operator $\hat{A}$, we define the Hermitian conjugate $\hat{A}^*$ by

$$\langle\phi|\hat{A}^*|\psi\rangle = \langle\psi|\hat{A}|\phi\rangle^*.$$

If $\hat{A}|\phi\rangle = |\chi\rangle$, then for any $|\psi\rangle$,

$$\langle\phi|\hat{A}^*|\psi\rangle = \langle\psi|\hat{A}|\phi\rangle^* = \langle\psi|\chi\rangle^* = \langle\chi|\psi\rangle,$$

so that we may write $\langle\phi|\hat{A}^* = \langle\chi|$. Observables correspond to Hermitian operators, such that $\hat{A}^* = \hat{A}$. For example, the operators defined by (10) are Hermitian, as may be proved by partial integration.

Every observable possesses a complete set of eigenvalues a and eigenvectors $|a\rangle$ such that

$$\hat{A}|a\rangle = a|a\rangle.$$

It is easy to prove that the eigenvalues a must be real, and that eigenvectors corresponding to different eigenvalues must be orthogonal. If the eigenvalues form a discrete set, then the corresponding eigenvectors can be normalized, so that

$$\langle a|a'\rangle = \delta_{aa'}. \tag{11}$$

The set of eigenvectors is complete in the sense that any vector $|\psi\rangle$ may be expanded as a linear combination of them,

$$|\psi\rangle = \sum_a |a\rangle \psi_a.$$

By (11), $\langle a|\psi\rangle = \psi_a$, so that we may also write the expansion in the form

$$|\psi\rangle = \sum_a |a\rangle\langle a|\psi\rangle$$

or, symbolically,

$$\hat{1} = \sum_a |a\rangle\langle a|. \tag{12}$$

Physically, $\langle a|\psi\rangle$ is related to the probability that the result of a measurement of $\hat{A}$ in the state $|\psi\rangle$ is a. In fact, this probability is $|\langle a|\psi\rangle|^2$ if $|\psi\rangle$ is normalized. The expectation value of $\hat{A}$ in the state $|\psi\rangle$ is

$$\bar{a} = \sum_a a|\langle a|\psi\rangle|^2 = \langle\psi|\hat{A}|\psi\rangle.$$

The uncertainty in the result of the measurement is characterized by the variance

$$\Delta a^2 = \overline{(a-\bar{a})^2} = \overline{(a^2)} - (\bar{a})^2.$$

If the set of eigenvalues is continuous, then instead of a sum in (12) we must have an integral,

$$\hat{1} = \int \mathrm{d}a\, |a\rangle\langle a|.$$

Correspondingly, the eigenvectors must be normalized according to

$$\langle a|a'\rangle = \delta(a-a'),$$

where δ is Dirac's delta function (mathematically a distribution), defined by

$$\int \mathrm{d}a'\, \delta(a-a')\, \mathrm{f}(a') = \mathrm{f}(a)$$

for any continuous function f. In this case, the individual eigenvectors are not normalizable, and do not really belong to the space of state-vectors. They do not correspond to real physical states, except in an idealized limiting sense. In this case, $|\langle a|\psi\rangle|^2 \mathrm{d}a$ is the probability that the result of the measurement of $\hat{A}$ lies between a and $a+\mathrm{d}a$.

The time development of a quantum-mechanical system is again governed by the Hamiltonian function, which is now an operator $\hat{H}$. It is described by Schrödinger's equation

$$i\hbar \frac{\partial}{\partial t}|\psi, t\rangle = \hat{H}|\psi, t\rangle. \tag{13}$$

If $|E\rangle$ is an eigenvector of $\hat{H}$, statisfying

$$\hat{H}|E\rangle = E|E\rangle,$$

then a particular solution of (13) is

$$|\psi, t\rangle = |E\rangle \exp(-iEt/\hbar).$$

In this case, the state-vector depends on time only through a phase factor, so that the physical state does not change with time. Since any state-vector can be expanded in terms of eigenvectors of $\hat{H}$, the general solution of (13) is a linear superposition of these stationary solutions.

$$|\psi, t\rangle = \exp(-i\hat{H}t/\hbar)|\psi, 0\rangle = \sum\!\!\!\!\!\!\int_{E} |E\rangle \exp(-iEt/\hbar)\langle E|\psi, 0\rangle, \tag{14}$$

where $\sum\!\!\!\!\!\!\int$ denotes a sum or integral, a sum over discrete eigenvalues and an integral over continuous eigenvalues.

1.3 THE HARMONIC OSCILLATOR

The quantum mechanical harmonic oscillator is described by operators $\hat{q}$ and $\hat{p}$ satisfying the canonical commutation relation,

$$[\hat{q}, \hat{p}] = i\hbar. \tag{15}$$

The Hamiltonian operator is (with no forcing term)

$$\hat{H} = \frac{1}{2m}\hat{p}^2 + \tfrac{1}{2}m\omega^2\hat{q}^2. \tag{16}$$

To find the eigenvalues of this operator, it is convenient to introduce a non-Hermitian operator $\hat{a}$ analogous to the classical complex variable z, but differently normalized, defined by

$$\hat{a} = \frac{m\omega\hat{q} + i\hat{p}}{(2\hbar m\omega)^{\frac{1}{2}}}. \tag{17}$$

From (15) it is easy to obtain the commutation relation

$$[\hat{a}, \hat{a}^*] = 1. \tag{18}$$

Substituting the inverse relations to (17),

$$\hat{q} = \left(\frac{\hbar}{2m\omega}\right)^{\frac{1}{2}}(\hat{a} + \hat{a}^*), \qquad \hat{p} = \left(\frac{\hbar m\omega}{2}\right)^{\frac{1}{2}}\frac{1}{i}(\hat{a} - \hat{a}^*) \tag{19}$$

into (16), we obtain

$$\hat{H} = \tfrac{1}{2}\hbar\omega(\hat{a}\hat{a}^* + \hat{a}^*\hat{a}) = \hbar\omega(\hat{a}^*\hat{a} + \tfrac{1}{2}) = (\hat{N} + \tfrac{1}{2})\hbar\omega, \tag{20}$$

where $\hat{N} = \hat{a}^*\hat{a}$.

Thus it suffices to find the eigenvalues of $\hat{N}$. If $|n\rangle$ is an eigenvector of $\hat{N}$, satisfying

$$\hat{N}|n\rangle = n|n\rangle, \tag{21}$$

then also

$$\hat{H}|n\rangle = E_n|n\rangle,$$

where

$$E_n = (n + \tfrac{1}{2})\hbar\omega. \tag{22}$$

Now from (18) we easily find that

$$[\hat{a}, \hat{N}] = \hat{a}\hat{a}^*\hat{a} - \hat{a}^*\hat{a}\hat{a} = [\hat{a}, \hat{a}^*]\hat{a} = \hat{a},$$

and similarly that

$$[\hat{a}^*, \hat{N}] = -\hat{a}^*.$$

Thus by (21)

$$\hat{N}\hat{a}|n\rangle = (\hat{a}\hat{N} - \hat{a})|n\rangle = (n-1)\hat{a}|n\rangle, \tag{23}$$

and

$$\hat{N}\hat{a}^*|n\rangle = (\hat{a}^*\hat{N} + \hat{a}^*)|n\rangle = (n+1)\hat{a}^*|n\rangle. \tag{24}$$

Thus $\hat{a}|n\rangle$ and $\hat{a}^*|n\rangle$ are also eigenvectors of $\hat{N}$, wtih eigenvalues $n-1$ and $n+1$, respectively. Starting from any eigenvalue n, we can go up and down a ladder of eigenvalues by unit steps. This ladder can only stop if, after some point, further application of $\hat{a}$ or $\hat{a}^*$ leads to a vector which vanishes. Now the norm of $\hat{a}|n\rangle$ is easily evaluated. Its square is

$$\langle n|\hat{a}^*\hat{a}|n\rangle = \langle n|\hat{N}|n\rangle = n. \tag{25}$$

Since this is a necessarily non-negative quantity, none of the eigenvalues can be negative. Moreover, it is zero only if $n = 0$. Thus, in order that the ladder should terminate before reaching negative eigenvalues, it must stop at $n = 0$, which means that the eigenvalues must be integers, $n = 0, 1, 2, \ldots$. Correspondingly, by (22), the eigenvalues of energy must be $\frac{1}{2}\hbar\omega, \frac{3}{2}\hbar\omega, \frac{5}{2}\hbar\omega, \ldots$. We have an equally spaced spectrum, with spacing $\hbar\omega$.

From (23) it follows that $\hat{a}|n\rangle$ must be proportional to the eigenvector $|n-1\rangle$, and by (25) the factor of proportionality must be $n^{\frac{1}{2}}$ multiplied by a phase factor, which can be chosen to be unity by a suitable choice of the arbitrary phases in the eigenvectors. Thus

$$\hat{a}|n\rangle = n^{\frac{1}{2}}|n-1\rangle. \tag{26}$$

To satisfy (21) we must then have also $\hat{a}^*|n-1\rangle = n^{\frac{1}{2}}|n\rangle$, or

$$\hat{a}^*|n\rangle = (n+1)^{\frac{1}{2}}|n+1\rangle. \tag{27}$$

As we have seen, the lowest eigenvalue E_0 of the Hamiltonian (16) is not zero, but $\frac{1}{2}\hbar\omega$. This "zero-point energy" can have observable consequences if we consider for example a system in which the frequency is variable. However, so long as we stick to a single oscillator of fixed frequency, it is merely an additive constant in the energy, and therefore unobservable. It will be convenient in what follows to subtract this constant, and consider the system defined by the Hamiltonian

$$\hat{H} = \frac{1}{2m}\hat{p}^2 + \tfrac{1}{2}m\omega^2\hat{q}^2 - \tfrac{1}{2}\hbar\omega = \hbar\omega\hat{a}^*\hat{a}. \tag{28}$$

Its eigenvalues are of course $E_n = n\hbar\omega$, $n = 0, 1, 2, \ldots$.

1.4 COHERENT STATES

We now wish to examine a particular set of states constructed out of the energy eigenstates of the harmonic oscillator. They have come to be known as "coherent" states, although the name is really more appropriate to the multimode case considered later than to the present single-mode system. They have properties which make them particularly convenient to use in discussions of the classical limit of the quantum oscillator.

For any complex number α, we define the state-vector

$$|\alpha\rangle = \sum_{n=0}^{\infty} |n\rangle \frac{\alpha^n}{(n!)^{\frac{1}{2}}} \exp(-\tfrac{1}{2}|\alpha|^2). \tag{29}$$

For $\alpha = 0$, this is simply the ground state $|0\rangle$, but for any other value of α, it is a state with some uncertainty in energy. The probability that the energy is E_n follows a Poisson distribution,

$$|\langle n|\alpha\rangle|^2 = \frac{|\alpha|^{2n}}{n!} \exp(-|\alpha|^2).$$

The scalar product of two such states is

$$\langle\alpha|\beta\rangle = \sum_{n=0}^{\infty} \frac{\alpha^{*n}\beta^n}{n!} \exp(-\tfrac{1}{2}|\alpha|^2) \exp(-\tfrac{1}{2}|\beta|^2) = \exp(\alpha^*\beta - \tfrac{1}{2}|\alpha|^2 - \tfrac{1}{2}|\beta|^2). \tag{30}$$

The states are therefore never orthogonal, although the scalar product is very small if $\alpha - \beta$ is large.

Now let us operate with the lowering operator $\hat{a}$ on a coherent state $|\alpha\rangle$. Using (26), and then replacing n by $n+1$ in the sum, we obtain

$$\hat{a}|\alpha\rangle = \sum_{n=1}^{\infty} n^{\frac{1}{2}}|n-1\rangle \frac{\alpha^n}{(n!)^{\frac{1}{2}}} \exp(-\tfrac{1}{2}|\alpha|^2)$$

$$= \sum_{n=0}^{\infty} |n\rangle \frac{\alpha^{n+1}}{(n!)^{\frac{1}{2}}} \exp(-\tfrac{1}{2}|\alpha|^2) = \alpha|\alpha\rangle. \tag{31}$$

The coherent states are therefore eigenstates of $\hat{a}$. (Recall that $\hat{a}$ is a non-Hermitian operator, and so can have complex eigenvalues, and non-orthogonal eigenvectors.) By conjugation, we also have $\langle\alpha|\hat{a}^* = \alpha^*\langle\alpha|$. It follows that the expectation value of any normally-ordered product (one in which all factors of $\hat{a}^*$ precede all factors of $\hat{a}$) in a coherent state is given by

$$\langle\alpha|\hat{a}^{*m}\hat{a}^n|\alpha\rangle = \alpha^{*m}\alpha^n.$$

Now also, since $\hat{a}|0\rangle = 0$, we have

$$\langle 0|(\hat{a}^* + \alpha^*)^m(\hat{a} + \alpha)^n|0\rangle = \alpha^{*m}\alpha^n.$$

More generally, any operator function $F(\hat{a}, \hat{a}^*)$ can be written, using the commutation relation (18), as a sum of normally-ordered products, and it follows that

$$\langle\alpha|F(\hat{a}, \hat{a}^*)|\alpha\rangle = \langle 0|F(\hat{a} + \alpha, \hat{a}^* + \alpha^*)|0\rangle. \tag{32}$$

In particular, it follows from (19) that

$$\langle\alpha|\hat{q}|\alpha\rangle = \left(\frac{2\hbar}{m\omega}\right)^{\frac{1}{2}} \mathrm{Re}\,\alpha, \qquad \langle\alpha|\hat{p}|\alpha\rangle = (2\hbar m\omega)^{\frac{1}{2}}\,\mathrm{Im}\,\alpha.$$

Moreover, since the uncertainties in $\hat{q}$ and $\hat{p}$ are unaffected by adding constant terms to $\hat{a}$ and $\hat{a}^*$, they are the same as in the ground state $|0\rangle$, namely

$$\Delta q^2 = \frac{\hbar}{2m\omega}, \qquad \Delta p^2 = \frac{\hbar m\omega}{2}.$$

Thus the product of uncertainties is

$$\Delta q\,\Delta p = \hbar/2,$$

which is the minimum value it can attain in any state.

Next, let us examine the time-development of a state which is initially a coherent state. If $|\psi, 0\rangle = |\alpha\rangle$, then from (14) we find

$$|\psi, t\rangle = \sum_n |n\rangle \exp(-in\omega t) \frac{\alpha^n}{(n!)^{\frac{1}{2}}} \exp(-\tfrac{1}{2}|\alpha|^2)$$

$$= \sum_n |n\rangle \frac{[\alpha \exp(-i\omega t)]^n}{(n!)^{\frac{1}{2}}} \exp(-\tfrac{1}{2}|\alpha|^2) = |\alpha \exp(-i\omega t)\rangle. \quad (33)$$

(If we had retained the original Hamiltonian (16) instead of the form (28) with the zero-point energy removed, we should have found an additional overall phase factor $\exp(-\frac{1}{2}i\omega t)$, which is of course of no physical significance.) We see from (33) that a state which is initially a coherent state is a coherent state at all times. The label α follows a circular path in the complex plane, described with constant angular velocity ω, exactly as in the solution of the classical oscillator. The uncertainties in $\hat{q}$ and $\hat{p}$ remain fixed, while their expectation values oscillate with the classical angular frequency ω. A coherent state is thus the quantum state which most closely approximates the motion of a classical oscillator. We note that the expectation value of the Hamiltonian (28) is

$$\langle\alpha|\hat{H}|\alpha\rangle = \hbar\omega|\alpha|^2,$$

which is exactly equal to the energy of a classical oscillator with the same amplitude. (In terms of the original Hamiltonian we would of course have in addition the zero-point energy term.)

A remarkable property of the coherent states is that any operator $\hat{A}$ is completely determined by its expectation values in all coherent states. For by (29),

$$\langle\alpha|\hat{A}|\alpha\rangle \exp|\alpha|^2 = \sum_{m,n} \frac{\alpha^{*m}\alpha^n}{(m!\, n!)^{\frac{1}{2}}} \langle m|\hat{A}|n\rangle,$$

so that the left hand side may be regarded as a generating function from which all the matrix elements of $\hat{A}$ between energy eigenstates can be recovered.

Thus to prove any operator equation it suffices to prove that its expectation values in all coherent states are satisfied. We may use this in particular to prove a completeness relation for the coherent states themselves. For any coherent state $|\beta\rangle$ we have, by (30),

$$\int \frac{d^2\alpha}{\pi} \langle\beta|\alpha\rangle \langle\alpha|\beta\rangle = \int \frac{d^2\alpha}{\pi} \exp(-|\alpha-\beta|^2) = 1 = \langle\beta|\beta\rangle,$$

where $d^2\alpha = d(\mathrm{Re}\,\alpha)\, d(\mathrm{Im}\,\alpha)$. Thus it follows that

$$\int \frac{d^2\alpha}{\pi} |\alpha\rangle \langle\alpha| = \hat{1}. \quad (34)$$

The coherent states in fact form an overcomplete family. They span the entire space of state-vectors, but are not linearly independent.

1.5 THE FORCED OSCILLATOR

The coherent states also play an important role in the quantum mechanical treatment of the forced oscillator, a problem which as we shall see later is essentially the same as that of an electromagnetic field in the presence of a prescribed classical current distribution.

The forced oscillator is described by the Hamiltonian

$$\hat{H} = \frac{1}{2m}\hat{p}^2 + \tfrac{1}{2}m\omega^2\hat{q}^2 - \tfrac{1}{2}\hbar\omega - \hat{q}\,\mathrm{f}(t) = \hbar\omega\hat{a}^*\hat{a} - \left(\frac{\hbar}{2m\omega}\right)^{\frac{1}{2}}(\hat{a}+\hat{a}^*)\,\mathrm{f}(t).$$

We know that in the absence of the forcing terms there are solutions of the Schrödinger equation in which the state is always a coherent state, and it is natural to ask whether this is still true of the forced oscillator. We therefore seek a solution in which $|\psi, t\rangle = |\alpha(t)\rangle \exp[i\phi(t)]$. Taking the scalar product of the Schrödinger equation with an arbitrary fixed coherent state $|\beta\rangle$, we have

$$i\hbar\frac{\partial}{\partial t}\Big\{\langle\beta|\alpha(t)\rangle \exp[i\phi(t)]\Big\} = \langle\beta|\hat{H}|\alpha(t)\rangle \exp[i\phi(t)]. \tag{35}$$

Clearly, it is sufficient (as well as necessary) to ensure that this equation is satisfied for arbitrary values of β.

Now,

$$\frac{\partial}{\partial t}\langle\beta|\alpha(t)\rangle = \frac{\partial}{\partial t}\exp[\beta^*\alpha(t) - \tfrac{1}{2}|\beta|^2 - \tfrac{1}{2}|\alpha(t)|^2]$$

$$= \langle\beta|\alpha(t)\rangle\left[\beta^*\frac{\mathrm{d}\alpha}{\mathrm{d}t} - \tfrac{1}{2}\frac{\mathrm{d}}{\mathrm{d}t}(|\alpha|^2)\right],$$

and

$$\langle\beta|\hat{H}|\alpha(t)\rangle = \langle\beta|\alpha(t)\rangle\left[\hbar\omega\beta^*\alpha - \left(\frac{\hbar}{2m\omega}\right)^{\frac{1}{2}}(\alpha+\beta^*)\,\mathrm{f}(t)\right].$$

Thus, substituting in (35), we obtain

$$i\hbar\left[\beta^*\frac{\mathrm{d}\alpha}{\mathrm{d}t} - \tfrac{1}{2}\frac{\mathrm{d}}{\mathrm{d}t}(|\alpha|^2)\right] - \hbar\frac{\mathrm{d}\phi}{\mathrm{d}t} = \hbar\omega\beta^*\alpha - \left(\frac{\hbar}{2m\omega}\right)^{\frac{1}{2}}(\alpha+\beta^*)\,\mathrm{f}(t).$$

This equation is self-consistent inasmuch as both sides are linear functions of β^* (in consequence of the linearity of the Hamiltonian in $\hat{a}^*$). We may equate

to zero the coefficients of β^* and 1, and thus obtain two equations for $d\alpha/dt$ and $d\phi/dt$. The first of these is essentially the same as the classical equation (6), namely

$$\frac{d\alpha}{dt} = -i\omega\alpha + \frac{i f(t)}{(2\hbar m\omega)^{\frac{1}{2}}}. \tag{36}$$

Using this equation, we can eliminate the term in $d(|\alpha|^2)/dt$ from the second equation, and obtain an equation for $d\phi/dt$ alone, namely

$$\frac{d\phi}{dt} = \frac{f(t)}{(2\hbar m\omega)^{\frac{1}{2}}} \operatorname{Re} \alpha(t). \tag{37}$$

These equations can be solved with any given initial conditions. Thus we see that if the system is initially in a coherent state, then at any later time it will again be in a coherent state.

In particular, let us suppose, as before, that f vanishes except during some finite time interval, $t_0 < t < t_1$, and that for $t < t_0$, the system is in its ground state $|0\rangle$. Then the solution of (36) is

$$\alpha(t) = \frac{i}{(2\hbar m\omega)^{\frac{1}{2}}} \int_{t_0}^{t} dt' f(t') \exp[-i\omega(t-t')],$$

as in (7). At any time $t > t_1$, the system will be in the state

$$|\psi, t\rangle = |\alpha_1 \exp(-i\omega t)\rangle \exp(i\phi_1), \tag{38}$$

where

$$\alpha_1 = \frac{i}{(2\hbar m\omega)^{\frac{1}{2}}} \int_{t_0}^{t_1} dt\, f(t) \exp(i\omega t) = \frac{i\tilde{f}(\omega)}{(2\hbar m\omega)^{\frac{1}{2}}} \tag{39}$$

and

$$\begin{aligned} \phi_1 &= \frac{1}{2\hbar m\omega} \int_{t_0}^{t_1} dt\, f(t) \int_{t_0}^{t} dt' f(t') \sin \omega(t-t') \\ &= \frac{1}{2\hbar m\omega} P \int_{-\infty}^{+\infty} \frac{d\omega'}{2\pi} \frac{|\tilde{f}(\omega')|^2}{\omega - \omega'}, \end{aligned}$$

where P denotes a principal value integral. By symmetry we can also write the solution as

$$\phi_1 = \frac{1}{\hbar m} P \int_{0}^{+\infty} \frac{d\omega'}{2\pi} \frac{|\tilde{f}(\omega')|^2}{\omega^2 - \omega'^2}. \tag{40}$$

We see therefore that the effect of applying an external force to a harmonic oscillator in its ground state is always to leave it in a coherent state, (38). By a suitable choice of the external force f(t), any given coherent state can be reached in this way.

2. Classical Electrodynamics

2.1 Potentials and Gauges

The basic equations of classical electrodynamics are Maxwell's equations, together with the Lorentz force equation

$$m\frac{\mathrm{d}\mathbf{v}}{\mathrm{d}t} = e\,[\mathbf{E}(\mathbf{r})+\mathbf{v}\wedge\mathbf{B}(\mathbf{r})], \qquad \mathbf{v} = \frac{\mathrm{d}\mathbf{r}}{\mathrm{d}t}\,, \tag{41}$$

which describes the motion of a particle of charge e and mass m in an electromagnetic field.

In rationalized units, and in the absence of dielectric or magnetic media, Maxwell's equations are

$$\nabla\,.\,\mathbf{B} = 0, \qquad \nabla\wedge\mathbf{E} + \frac{\partial\mathbf{B}}{\partial t} = \mathbf{0} \tag{42}$$

and

$$\varepsilon_0\nabla\,.\,\mathbf{E} = \rho, \qquad \mu_0{}^{-1}\,\nabla\wedge\mathbf{B} - \varepsilon_0\frac{\partial\mathbf{E}}{\partial t} = \mathbf{j}, \tag{43}$$

where ρ and $\mathbf{j}$ are the electric charge and current densities.

From (43) there follows immediately the continuity equation

$$\frac{\partial\rho}{\partial t} + \nabla\,.\,\mathbf{j} = 0 \tag{44}$$

which expresses the microscopic conservation of charge.

From (42) we may infer the existence of a vector potential $\mathbf{A}$ and a scalar potential ϕ such that

$$\mathbf{B} = \nabla\wedge\mathbf{A}, \qquad \mathbf{E} = -\frac{\partial\mathbf{A}}{\partial t} - \nabla\phi. \tag{45}$$

These potentials are not unique. The fields **E** and **B** are unchanged if we make any gauge transformation,

$$\mathbf{A}\rightarrow\mathbf{A}-\nabla\lambda, \qquad \phi\rightarrow\phi+\frac{\partial\lambda}{\partial t}, \tag{46}$$

where λ is any function of space and time, This arbitrariness may be removed by imposing a gauge condition. For our purposes, the most convenient choice is the radiation gauge, defined by the condition

$$\nabla\,.\,\mathbf{A}=0. \tag{47}$$

If we start with any potentials satisfying (45), then we can pass to the radiation-gauge potentials by making a gauge transformation (46) in which the function λ is chosen to be a solution of $\nabla^2\lambda=\nabla\,.\,\mathbf{A}$.

If we substitute for **E** from (45) into the first of the equations (43), and use the radiation-gauge condition (47), we obtain

$$-\varepsilon_0\nabla^2\phi=\rho.$$

If the charge density ρ vanishes outside some finite region of space (or at least tends rapidly to zero as $r\rightarrow\infty$), then we can solve this equation explicitly in the form

$$\phi(\mathbf{r})=\int \mathrm{d}^3\mathbf{r}'\,\frac{\rho(\mathbf{r}')}{4\pi\varepsilon_0|\mathbf{r}-\mathbf{r}'|}.$$

We shall write this equation symbolically as

$$\phi=-\frac{1}{\varepsilon_0\nabla^2}\,\rho. \tag{48}$$

Thus the radiation-gauge scalar potential is just the usual potential of electrostatics.

Using (48) we may eliminate ϕ and obtain an equation for **A** alone. When we substitute from (45) into the second of the equations (43), we obtain a term

$$\varepsilon_0\nabla\frac{\partial\phi}{\partial t}=-\nabla\frac{1}{\nabla^2}\,\frac{\partial\rho}{\partial t}=\nabla\frac{1}{\nabla^2}\nabla\,.\,\mathbf{j},$$

by (44). Hence, using the standard vector identity

$$\nabla\wedge(\nabla\wedge\mathbf{A})=\nabla\nabla\,.\,\mathbf{A}-\nabla^2\mathbf{A},$$

and the radiation-gauge condition (47), we obtain

$$\varepsilon_0 \frac{\partial^2 \mathbf{A}}{\partial t^2} - \mu_0{}^{-1} \nabla^2 \mathbf{A} = \mathbf{j} - \nabla \frac{1}{\nabla^2} \nabla \cdot \mathbf{j} = \mathbf{j}_T, \tag{49}$$

say. The right hand side of this equation is the divergenceless, or "transverse", part of $\mathbf{j}$, satisfying $\nabla \cdot \mathbf{j}_T = 0$.

When $\mathbf{j}$ is identically zero, $\mathbf{A}$ satisfies the wave equation, which describes waves propagating with velocity $c = (\varepsilon_0 \mu_0)^{-\frac{1}{2}}$. A particular solution of this equation is a plane wave,

$$\mathbf{A}(\mathbf{r}, t) = \mathrm{Re}\,[(\mathbf{A}_0 \exp(i\mathbf{k} \cdot \mathbf{r} - i\omega_k t)], \tag{50}$$

where $\omega_k = c|\mathbf{k}|$, and $\mathbf{A}_0$ is a constant complex vector satisfying $\mathbf{k} \cdot \mathbf{A}_0 = 0$. The corresponding electric and magnetic fields are given by

$$\begin{aligned} \mathbf{E}(\mathbf{r}, t) &= \mathrm{Re}\,[i\omega_k \mathbf{A}_0 \exp(i\mathbf{k} \cdot \mathbf{r} - i\omega_k t)], \\ \mathbf{B}(\mathbf{r}, t) &= \mathrm{Re}\,[i\mathbf{k} \wedge \mathbf{A}_0 \exp(i\mathbf{k} \cdot \mathbf{r} - i\omega_k t)]. \end{aligned} \tag{51}$$

The general solution is of course a linear superposition of these.

From Maxwell's equations (42) and (43) we find

$$\frac{\partial}{\partial t}(\tfrac{1}{2}\varepsilon_0 \mathbf{E}^2 + \tfrac{1}{2}\mu_0{}^{-1} \mathbf{B}^2) = \mathbf{E} \cdot (\mu_0{}^{-1} \nabla \wedge \mathbf{B} - \mathbf{j}) - \mu_0{}^{-1} \mathbf{B} \cdot \nabla \wedge \mathbf{E}.$$

Using the standard vector identity

$$\nabla \cdot (\mathbf{a} \wedge \mathbf{b}) = \mathbf{b} \cdot \nabla \wedge \mathbf{a} - \mathbf{a} \cdot \nabla \wedge \mathbf{b}, \tag{52}$$

we may write this in the form

$$\frac{\partial u}{\partial t} + \nabla \cdot \mathbf{S} = -\mathbf{E} \cdot \mathbf{j}, \tag{53}$$

where

$$u = \tfrac{1}{2}(\varepsilon_0 \mathbf{E}^2 + \mu_0{}^{-1} \mathbf{B}^2) \tag{54}$$

and

$$\mathbf{S} = \mu_0{}^{-1} \mathbf{E} \wedge \mathbf{B}. \tag{55}$$

The right hand side of (53) is the negative of the rate at which the electric field does work on the currents per unit volume. Thus we may interpret (53) as the continuity equation for energy, if we identify u with the energy density in the field, and $\mathbf{S}$ with the energy flux vector (Poynting vector). In particular, the total energy stored in the field is,

$$H_I = \int \mathrm{d}^3\mathbf{r}\, \tfrac{1}{2}(\varepsilon_0 \mathbf{E}^2 + \mu_0{}^{-1} \mathbf{B}^2). \tag{56}$$

It may be noted that the contribution to (56) which comes from the "longitudinal" part $-\nabla\phi$ of $\mathbf{E}$ can be cast by partial integration (Green's theorem) into the form

$$-\tfrac{1}{2}\int \mathrm{d}^3\mathbf{r}\varepsilon_0\phi\nabla^2\phi = \tfrac{1}{2}\int \mathrm{d}^3\mathbf{r}\,\phi\rho = \tfrac{1}{2}\int \mathrm{d}^3\mathbf{r}\int \mathrm{d}^3\mathbf{r}'\,\frac{\rho(\mathbf{r})\,\rho(\mathbf{r}')}{4\pi\varepsilon_0|\mathbf{r}-\mathbf{r}'|}. \tag{57}$$

This is the electrostatic energy corresponding to the charge distribution ρ.

We note also that for a plane wave (51) the energy density and energy flux vector, averaged over a period of oscillation, are

$$\langle u\rangle_{av} = I/c, \qquad \langle \mathbf{S}\rangle_{av} = I\mathbf{k}, \tag{58}$$

where the intensity I is given by

$$I = \tfrac{1}{2}c\varepsilon_0\omega_k{}^2\mathbf{A}_0{}^*\,.\,\mathbf{A}_0. \tag{59}$$

2.2 HAMILTONIAN FORM OF EQUATIONS OF MOTION

We now wish to show that the equation of motion (41) for a charged particle may be derived from the Hamiltonian function

$$H = \frac{1}{2m}[\mathbf{p}-e\mathbf{A}(\mathbf{r},t)]^2 + e\phi(\mathbf{r},t). \tag{60}$$

The first set of Hamilton's equations is

$$\mathbf{v} = \frac{\mathrm{d}\mathbf{r}}{\mathrm{d}t} = \frac{\partial H}{\partial \mathbf{p}} = \frac{1}{m}(\mathbf{p}-e\mathbf{A}), \tag{61}$$

where we have used the notation $\partial/\partial\mathbf{p}$ for the gradient operator with respect to the momentum vector $\mathbf{p}$. Since $\mathbf{r}$ appears in H only through the potentials $\mathbf{A}$ and ϕ, the second set is

$$\frac{\mathrm{d}\mathbf{p}}{\mathrm{d}t} = -\frac{\partial H}{\partial \mathbf{r}} = \frac{e}{m}(\nabla\mathbf{A})\cdot(\mathbf{p}-e\mathbf{A}) - e\nabla\phi,$$

which may be written

$$\frac{\mathrm{d}\mathbf{p}}{\mathrm{d}t} = e(\nabla\mathbf{A})\,.\,\mathbf{v} - e\nabla\phi. \tag{62}$$

Now, since $\mathbf{A}$ depends on t both explicitly and through its dependence on $\mathbf{r}$,

we have

$$\frac{\mathrm{d}\mathbf{A}}{\mathrm{d}t} = \frac{\partial \mathbf{A}}{\partial t} + \mathbf{v} \cdot \nabla \mathbf{A}. \tag{63}$$

Hence, differentiating (61) with respect to the time, and using (62) and (63), we obtain

$$\begin{aligned} m\frac{\mathrm{d}\mathbf{v}}{\mathrm{d}t} = \frac{\mathrm{d}\mathbf{p}}{\mathrm{d}t} - e\frac{\mathrm{d}\mathbf{A}}{\mathrm{d}t} &= e(\nabla \mathbf{A}) \cdot \mathbf{v} - e\nabla\phi - e\frac{\partial \mathbf{A}}{\partial t} - e\mathbf{v} \cdot \nabla \mathbf{A} \\ &= e\left(-\nabla\phi - \frac{\partial \mathbf{A}}{\partial t}\right) + e\mathbf{v} \wedge (\nabla \wedge \mathbf{A}) \\ &= e(\mathbf{E} + \mathbf{v} \wedge \mathbf{B}), \end{aligned}$$

by (45). Thus we have shown that the Hamiltonian (60) correctly reproduces the equations of motion (41).

We note that the first term in H is numerically equal to the kinetic energy of the particle, $\frac{1}{2}m\mathbf{v}^2$, while the second term is the electrostatic potential energy. This second term is of course included in (57). Thus to obtain the total Hamiltonian function for the system comprising both the electromagnetic field and the particles, this term in (60) should be omitted if it is included in (56).

3. Semiclassical Electrodynamics

3.1 Perturbation Theory

Before going on to discuss semiclassical electrodynamics, it will be useful to review briefly the elements of quantum-mechanical perturbation theory, in its time-dependent form.

We suppose that the Hamiltonian operator may be written in the form

$$\hat{H} = \hat{H}_0 + \hat{H}_1(t), \tag{64}$$

where $\hat{H}_1(t)$ may be treated as a small perturbation, and where it is assumed that we already know the eigenvalues E_n and eigenvectors $|n\rangle$ of $\hat{H}_0$, satisfying

$$\hat{H}_0|n\rangle = E_n|n\rangle. \tag{65}$$

We then seek a solution of the Schrödinger equation for given initial conditions as a series in powers of $\hat{H}_1$.

To do this, we expand the solution $|\psi, t\rangle$ of the Schrödinger equation in terms of the eigenvectors $|n\rangle$,

$$|\psi, t\rangle = \sum_n |n\rangle\, c_n(t).$$

Using (65) we may write the Schrödinger equation as

$$i\hbar\sum_n |n\rangle \frac{dc_n}{dt} = \sum_n E_n|n\rangle\, c_n(t) + \sum_n \hat{H}_1(t)\,|n\rangle\, c_n(t).$$

Then, taking the scalar product with an eigenstate $|m\rangle$ and using the orthonormality conditions (11), we obtain

$$i\hbar \frac{dc_m}{dt} = E_m\, c_m(t) + \sum_n \langle m|\hat{H}_1(t)\,|n\rangle\, c_n(t).$$

It is convenient to extract the time dependence of the unperturbed system by setting

$$c_n(t) = u_n(t) \exp(-iE_n t/\hbar),$$

so that the equation becomes

$$i\hbar \frac{du_m}{dt} = \sum_n \langle m|\hat{H}_1(t)\,|n\rangle \exp(i\omega_{mn} t)\, u_n(t) \tag{66}$$

where $\omega_{mn} = (E_m - E_n)/\hbar$.

Now let us suppose that initially, at $t = 0$, the system is in one of its eigenstates, $|\psi, 0\rangle = |n\rangle$, say. Equivalently, $u_m(0) = \delta_{mn}$. We take this as our zeroth order solution for all t,

$$u_m^{(0)}(t) = \delta_{mn}.$$

Then the complete solution may be expressed as a series

$$u_m(t) = \sum_r u_m^{(r)}(t),$$

where higher terms are obtained by iteration from (66). Explicitly, $u^{(r)}$ is obtained by solving the equation with u replaced by $u^{(r-1)}$ on the right, which yields

$$u_m^{(r)}(t) = \frac{1}{i\hbar} \sum_n \int_0^t dt' \langle m|\hat{H}_1(t')\,|n\rangle \exp(i\omega_{mn} t')\, u_n^{(r-1)}(t'). \tag{67}$$

We shall consider explicitly only the first-order solution

$$u_m^{(1)}(t) = \frac{1}{i\hbar} \int_0^t dt' \langle m|\hat{H}_1(t')\,|n\rangle \exp(i\omega_{mn} t'). \tag{68}$$

To be specific, let us suppose that $\hat{H}_1$ is periodic in time, with the form

$$\hat{H}_1(t) = \hat{V} \exp(-i\omega t) + \hat{V}^* \exp(i\omega t). \tag{69}$$

Then we can perform the integrals and obtain

$$u_m^{(1)}(t) = \langle m|\hat{V}|n\rangle \frac{1-\exp[i(\omega_{mn}-\omega)t]}{\hbar(\omega_{mn}-\omega)} + \langle m|\hat{V}^*|n\rangle \frac{1-\exp[i(\omega_{mn}+\omega)t]}{\hbar(\omega_{mn}+\omega)} \tag{70}$$

Because of the presence of the denominators, the transition probability from state $|n\rangle$ to state $|m\rangle$ can be large only if $\omega_{mn} \simeq \pm\omega$. Let us suppose that $\omega_{mn} \simeq \omega$, so that the second term in (70) is negligible. Then the transition probability per unit time to state $|m\rangle$ is, in the limit of long time,

$$\begin{aligned} w_{mn} &= \lim_{t\to\infty} (1/t)\,|u_m^{(1)}(t)|^2 \\ &= |\langle m|\hat{V}|n\rangle|^2 \lim_{t\to\infty} \frac{1}{t} \left|\frac{1-\exp[i(\omega_{mn}-\omega)t]}{\hbar(\omega_{mn}-\omega)}\right|^2. \end{aligned}$$

Now clearly if $\omega_{mn} \neq \omega$, then the limit is zero, because the squared modulus is always finite. On the other hand, for $\omega_{mn} = \omega$, the quantity inside the modulus sign depends linearly on t, so that the limit is infinite. The limit therefore behaves like a Dirac delta function, $\delta(\omega_{mn}-\omega)$. We note that

$$\int_{-\infty}^{+\infty} d\omega \frac{1}{t}\left|\frac{1-\exp[i(\omega_{mn}-\omega)\,t]}{\hbar(\omega_{mn}-\omega)}\right|^2 = \frac{2}{\hbar^2}\int_{-\infty}^{+\infty} dx \frac{1-\cos x}{x^2} = \frac{2\pi}{\hbar^2},$$

independent of t. Hence the integrand must tend to the limit $(2\pi/\hbar^2)\delta(\omega_{mn}-\omega)$. Thus we obtain for the transition probability per unit time the expression

$$w_{mn} = \frac{2\pi}{\hbar^2}|\langle m|\hat{V}|n\rangle|^2\,\delta(\omega_{mn}-\omega) = \frac{2\pi}{\hbar}|\langle m|\hat{V}|n\rangle|^2\,\delta(E_m-E_n-\hbar\omega). \tag{71}$$

If we have a strictly periodic perturbation, and discrete energy levels, we can never get a finite transition probability. To obtain a finite result we have either to integrate over a range of values of the final-state energy, or over a range of values of the perturbing potential.

We can calculate similarly the transition probability for a downward transition, using the second term of (70). The result is

$$w_{mn} = \frac{2\pi}{\hbar^2}|\langle m|\hat{V}^*|n\rangle|^2\,\delta(\omega_{mn}+\omega) = \frac{2\pi}{\hbar}|\langle m|\hat{V}^*|n\rangle|^2\,\delta(E_m-E_n+\hbar\omega). \tag{72}$$

Comparing these two expressions, (71) and (72), we see that $w_{mn} = w_{nm}$ always. This is an expression of the principle of detailed balance.

3.2 SEMICLASSICAL TREATMENT OF INDUCED TRANSITIONS

In semiclassical theory, we treat the electrons quantum mechanically, but regard the electromagnetic field as an externally prescribed classical field. The Hamiltonian operator for an electron in an atom, and in the presence of an external radiation field is obtained from (60) by replacing $\mathbf{r}$ and $\mathbf{p}$ by operators $\hat{\mathbf{r}}$ and $\hat{\mathbf{p}}$. It is

$$\hat{H} = \frac{1}{2m}[\hat{\mathbf{p}} - e\mathbf{A}(\hat{\mathbf{r}}, t)]^2 + V(\hat{\mathbf{r}}), \tag{73}$$

where $V(\hat{\mathbf{r}})$ is the (Coulomb) potential responsible for atomic binding. We may write this Hamiltonian in the form (64) with

$$\hat{H}_0 = \frac{1}{2m}\hat{\mathbf{p}}^2 + V(\hat{\mathbf{r}})$$

and

$$\hat{H}_1(t) = -\frac{e}{2m}[\hat{\mathbf{p}} \cdot \mathbf{A}(\hat{\mathbf{r}}, t) + \mathbf{A}(\hat{\mathbf{r}}, t) \cdot \hat{\mathbf{p}}] + \frac{e^2}{2m}\mathbf{A}(\hat{\mathbf{r}}, t)^2.$$

The difference between the two terms in the square brackets is

$$\hat{\mathbf{p}} \cdot \hat{\mathbf{A}} - \hat{\mathbf{A}} \cdot \hat{\mathbf{p}} = -i\hbar \nabla \cdot \mathbf{A} = 0$$

in the radiation gauge. Moreover, if we work only to first order in e, then the last term in H_1 is negligible, and we may take

$$\hat{H}_1(t) = -\frac{e}{m}\hat{\mathbf{p}} \cdot \mathbf{A}(\hat{\mathbf{r}}, t). \tag{74}$$

Let us suppose that the external field is a plane wave, of the form (50). Then the perturbation (74) is periodic in t, and can be expressed in the form (69), with $\omega = \omega_k$ and

$$\hat{V} = -\frac{e}{2m}\hat{\mathbf{p}} \cdot \mathbf{A}_0 \exp(i\mathbf{k} \cdot \hat{\mathbf{r}}).$$

Thus according to (71) the transition probability per unit time for an induced absorption process is

$$w_{mn} = \frac{2\pi}{\hbar^2}\left|\frac{e}{2m}\mathbf{A}_0 \cdot \langle m|\hat{\mathbf{p}} \exp(i\mathbf{k} \cdot \hat{\mathbf{r}})|n\rangle\right|^2 \delta(\omega_{mn} - \omega_k).$$

For optical frequencies, the wavelength of the electromagnetic wave is long compared with the atomic dimensions. Thus it is a good approximation to

replace the factor $\exp(i\mathbf{k}.\mathbf{r})$ in the matrix element by 1. This is the dipole approximation. Using the relation

$$[\hat{\mathbf{r}}, \hat{H}_0] = \frac{i\hbar}{m}\hat{\mathbf{p}},$$

we can obtain a relation between the matrix elements of $\hat{\mathbf{p}}$ and of $\hat{\mathbf{r}}$, namely

$$\frac{i\hbar}{m}\langle m|\hat{\mathbf{p}}|n\rangle = \langle m|[\hat{\mathbf{r}}, \hat{H}_0]|n\rangle = (E_n - E_m)\langle m|\hat{\mathbf{r}}|n\rangle = -\hbar\omega_{mn}\langle m|\hat{\mathbf{r}}|n\rangle.$$

Hence we find

$$w_{mn} = \frac{2\pi}{\hbar^2}\frac{e^2}{4}\omega_k{}^2\,\mathbf{A}_0{}^*.\mathbf{A}_0|\mathbf{a}.\langle m|\hat{\mathbf{r}}|n\rangle|^2\delta(\omega_{mn}-\omega_k), \tag{75}$$

where $\mathbf{a}$ is the complex unit vector in the direction of $\mathbf{A}_0$, $\mathbf{a} = \mathbf{A}_0/(\mathbf{A}_0{}^*.\mathbf{A}_0)^{\frac{1}{2}}$. As expected, the transition probability is proportional to the intensity I given by (59).

If we suppose that the field is an incoherent mixture of components with different frequencies, then we have to sum over all frequencies. If the intensity in the frequency range between ω and $\omega+\mathrm{d}\omega$ is $I(\omega)\mathrm{d}\omega$, then the total transition probability per unit time is

$$w_{mn} = (2\pi)^2\frac{\alpha}{\hbar}I(\omega_{mn})|\mathbf{a}.\langle m|\hat{\mathbf{r}}|n\rangle|^2, \tag{76}$$

where α is the fine structure constant, a dimensionless number,

$$\alpha = \frac{e^2}{4\pi\varepsilon_0\hbar c} \simeq \frac{1}{137}. \tag{77}$$

This discussion covers only induced transitions, not spontaneous ones, which can only be treated correctly in the fully quantized theory.

4. Quantization of the Electromagnetic Field

4.1 Mode decomposition of the electromagnetic field

In order to quantize the electromagnetic field, it is convenient first to decompose it into modes.

Let us begin by considering the field enclosed in a cavity of volume, V, and satisfying prescribed boundary conditions on the walls. For example, we may

choose the walls to be perfectly conducting, which requires that the electric field must be normal to the walls,

$$\mathbf{n} \wedge \mathbf{E} = \mathbf{0} \tag{78}$$

on the boundaries, where $\mathbf{n}$ is the unit normal.

In the cavity (where for the moment we assume there are no charges or currents), the radiation-gauge vector potential satisfies the wave equation

$$\varepsilon_0 \frac{\partial^2 \mathbf{A}}{\partial t^2} - \mu_0{}^{-1} \nabla^2 \mathbf{A} = \mathbf{0}, \tag{79}$$

together with the appropriate boundary conditions, for example

$$\mathbf{n} \wedge \mathbf{A} = \mathbf{0}. \tag{80}$$

We now look for periodic solutions of (79). To do this we have to find all values of k^2 for which the equations

$$(\nabla^2 + k^2)\,\mathbf{f}_k = \mathbf{0}, \qquad \nabla \cdot \mathbf{f}_k = 0 \tag{81}$$

have solutions satisfying the boundary conditions, say

$$\mathbf{n} \wedge \mathbf{f}_k = \mathbf{0}. \tag{82}$$

If the subscript k labels the set of all such solutions, then we can expand the general solution of (79) in terms of these functions, in the form

$$\mathbf{A}(\mathbf{r}, t) = \sum_k q_k(t)\,\mathbf{f}_k(\mathbf{r}), \tag{83}$$

where $q_k(t)$ satisfies the equation

$$\frac{\mathrm{d}^2 q_k}{\mathrm{d}t^2} + \omega_k{}^2 q_k = 0, \tag{84}$$

with $\omega_k{}^2 = c^2 k^2$.

The corresponding magnetic field, given by (45), is

$$\mathbf{B}(\mathbf{r}, t) = \sum_k q_k(t)\,\nabla \wedge \mathbf{f}_k(\mathbf{r}) \tag{85}$$

while the electric field may be written

$$\mathbf{E}(\mathbf{r}, t) = -\sum_k p_k(t)\,\mathbf{f}_k(\mathbf{r}), \tag{86}$$

where $p_k = \mathrm{d}q_k/\mathrm{d}t$. With this definition, we may write (84) in the form

$$\frac{\mathrm{d}q_k}{\mathrm{d}t} = p_k, \qquad \frac{\mathrm{d}p_k}{\mathrm{d}t} = -\omega_k{}^2 q_k. \tag{87}$$

These are the equations, in Hamiltonian form, of a set of harmonic oscillators of unit mass and angular frequency ω_k. Thus the electromagnetic field has been decomposed into modes, each behaving like a harmonic oscillator.

It may be anticipated that, with suitable normalization of the mode functions $\mathbf{f}_k(\mathbf{r})$, the Hamiltonian (56) of the electromagnetic field can be written as a sum of harmonic oscillator Hamiltonians. To show that this is indeed the case, we have first to establish certain orthogonality properties of the mode functions.

Using the vector identity (52) with $\mathbf{a} = \mathbf{f}_l$ and $\mathbf{b} = \nabla \wedge \mathbf{f}_k$, we have

$$\nabla . [\mathbf{f}_l \wedge (\nabla \wedge \mathbf{f}_k)] = (\nabla \wedge \mathbf{f}_k) . (\nabla \wedge \mathbf{f}_l) - \mathbf{f}_l . [\nabla \wedge (\nabla \wedge \mathbf{f}_k)]. \tag{88}$$

We now integrate over the volume V, and use Green's theorem to express the left hand side as a surface integral over the boundary S of V,

$$\int_S \mathbf{dS} . \mathbf{f}_l \wedge (\nabla \wedge \mathbf{f}_k) = \int_S (\mathbf{dS} \wedge \mathbf{f}_l) . (\nabla \wedge \mathbf{f}_k),$$

which vanishes in virtue of the boundary condition (82). On the other hand, by (81),

$$\nabla \wedge (\nabla \wedge \mathbf{f}_k) = \nabla \nabla . \mathbf{f}_k - \nabla^2 \mathbf{f}_k = k^2 \mathbf{f}_k.$$

Hence we obtain

$$\int_V \mathrm{d}^3\mathbf{r}\, (\nabla \wedge \mathbf{f}_k) . (\nabla \wedge \mathbf{f}_l) = k^2 \int_V \mathrm{d}^3\mathbf{r}\, \mathbf{f}_k . \mathbf{f}_l = l^2 \int_V \mathrm{d}^3\mathbf{r}\, \mathbf{f}_k . \mathbf{f}_l, \tag{89}$$

where the last equality follows by symmetry. Thus the integrals vanish unless $k^2 = l^2$. We shall see in a moment that the correct normalization to choose is

$$\varepsilon_0 \int_V \mathrm{d}^3\mathbf{r}\, \mathbf{f}_k . \mathbf{f}_l = \delta_{kl}. \tag{90}$$

For then the first term in the Hamiltonian (56) becomes

$$\tfrac{1}{2}\varepsilon_0 \int_V \mathrm{d}^3\mathbf{r}\, \mathbf{E}^2 = \tfrac{1}{2} \sum_k \sum_l p_k p_l\, \varepsilon_0 \int_V \mathrm{d}^3\mathbf{r}\, \mathbf{f}_k . \mathbf{f}_l = \tfrac{1}{2} \sum_k p_k^2,$$

while the second may be written, using (89) and (90), as

$$\tfrac{1}{2}\mu_0^{-1} \int_V \mathrm{d}^3\mathbf{r}\, \mathbf{B}^2 = \tfrac{1}{2} \sum_k \sum_l q_k q_l\, \mu_0^{-1} \int \mathrm{d}^3\mathbf{r}\, (\nabla \wedge \mathbf{f}_k) . (\nabla \wedge \mathbf{f}_l) = \tfrac{1}{2} \sum_k \omega_k^2 q_k^2.$$

Thus the total Hamiltonian of the electromagnetic field is

$$H_F = \sum_k \tfrac{1}{2}(p_k^2 + \omega_k^2 q_k^2), \tag{91}$$

in agreement with (87).

4.2 FIELD QUANTIZATION

Now that we have reduced the Hamiltonian for the electromagnetic field to a sum of harmonic-oscillator Hamiltonians, the quantization of the system is straightforward. We have to replace the classical variables q_k and p_k by operators $\hat{q}_k$ and $\hat{p}_k$ satisfying the canonical commutation relations (9). We choose the Hamiltonian of each oscillator in the form (28) with the zero-point energy removed, and therefore obtain for the total Hamiltonian the expression

$$\hat{H}_F = \sum_k \tfrac{1}{2}(\hat{p}_k^2 + \omega_k^2 \hat{q}_k^2 - \hbar\omega_k) = \sum_k \hbar\omega_k \hat{a}_k^* \hat{a}_k, \tag{92}$$

where as before we have defined the non-Hermitian operator

$$\hat{a}_k = \frac{\omega_k \hat{q}_k + i\hat{p}_k}{(2\hbar\omega_k)^{\frac{1}{2}}}. \tag{93}$$

It may be noted that the removal of the zero-point energy is in this case essential if we want to have finite energies, because the sum over all modes of the zero-point energy,

$$\sum_k \tfrac{1}{2}\hbar\omega_k,$$

is infinite. The only observable energies are of course energy differences between states, and these are finite. The Hamiltonian (92) is chosen so that its lowest eigenvalue, the energy of the ground state or vacuum state, is zero.

The ladder operators (93) satisfy the commutation relations

$$[\hat{a}_k, \hat{a}_l] = [\hat{a}_k^*, \hat{a}_l^*] = 0, \qquad [\hat{a}_k, \hat{a}_l^*] = \delta_{kl}. \tag{94}$$

The ground state $|0\rangle$ is characterized by the condition that

$$\hat{a}_k|0\rangle = 0 \tag{95}$$

for all k. Other energy eigenstates may be obtained by acting with the operators $\hat{a}_k^*$ successively. For example,

$$|k\rangle = \hat{a}_k^*|0\rangle. \tag{96}$$

is an eigenstate of energy $\hbar\omega_k$, which may be interpreted as a state containing one photon in the kth mode, and

$$|k, l\rangle = \hat{a}_k^* \hat{a}_l^*|0\rangle \tag{97}$$

is an eigenstate of energy $\hbar(\omega_k+\omega_l)$, a two-photon state with one photon in each of the two modes labelled by k and l. (If $k = l$, it is a state with two photons in the kth mode. In that case, it should have an additional normalization factor $1/2^{\frac{1}{2}}$.)

In general, each energy eigenstate may be labelled by a set of numbers $n_1, n_2, \ldots$, one for each mode. The number n_k represents the energy level of the kth mode, or equivalently the number of photons in that mode. Each n_k may range over the values 0, 1, 2, ..., but only a finite number of them may be different from zero. Denoting the set of all the numbers n_k by $\{n\}$, we may write this eigenvector as

$$|\{n\}\rangle = |n_1, n_2, \ldots, n_k, \ldots\rangle.$$

The corresponding energy eigenvalue is

$$E_{\{n\}} = \sum_k E_{n_k} = \sum_k n_k \hbar\omega_k. \tag{98}$$

This state is of course an eigenstate of each of the operators $\hat{N}_k = \hat{a}_k^* \hat{a}_k$, with eigenvalue n_k, and also of the total photon number operator

$$\hat{N} = \sum_k \hat{N}_k$$

with eigenvalue

$$n = \sum_k n_k.$$

The effect of any one of the operators $\hat{a}_k$ or $\hat{a}_k^*$ on a state $|\{n\}\rangle$ is to change the value of the label n_k by one unit, while leaving all the other labels unaltered:

$$\begin{aligned} \hat{a}_k|\{n\}\rangle &= (n_k)^{\frac{1}{2}}|n_1, n_2, \ldots, n_k-1, \ldots\rangle, \\ \hat{a}_k^*|\{n\}\rangle &= (n_k+1)^{\frac{1}{2}}|n_1, n_2, \ldots, n_k+1, \ldots\rangle \end{aligned} \tag{99}$$

Their effect is to remove one photon, or to add one extra photon, and they are therefore called annihilation and creation operators respectively.

Just as for a single oscillator, we can again define coherent states. Each coherent state is labelled by a set of complex numbers $\{\alpha\} = \{\alpha_1, \alpha_2, \ldots\}$. It is defined by

$$|\{\alpha\}\rangle = \sum_{n_1, n_2, \ldots} |\{n\}\rangle \prod_k \frac{\alpha_k^{n_k}}{(n_k!)^{\frac{1}{2}}} \exp(-\tfrac{1}{2}|\alpha_k|^2). \tag{100}$$

Using (99), it is easy to prove, as before, that these states are eigenstates of each of the annihilation operators $\hat{a}_k$. They satisfy

$$\hat{a}_k|\{\alpha\}\rangle = \alpha_k|\{\alpha\}\rangle, \tag{101}$$

and hence also

$$\langle\{\alpha\}|\,\hat{a}_k^* = \alpha_k^*\,\langle\{\alpha\}|. \tag{102}$$

It follows from these relations that

$$\langle\{\alpha\}|\hat{N}_k|\{\alpha\}\rangle = \langle\{\alpha\}|\hat{a}_k^*\,\hat{a}_k|\{\alpha\}\rangle = |\alpha_k|^2.$$

Thus, in order to ensure that the expectation value of the total photon number is finite, we must impose on the set of numbers α_k the condition

$$\langle\{\alpha\}|\hat{N}|\{\alpha\}\rangle = \sum_k |\alpha_k|^2 < \infty. \tag{103}$$

(This is also necessary to ensure that the product of the exponential factors in (100) is convergent.)

The scalar product of two coherent states is easily evaluated, and is given by

$$\langle\{\alpha\}|\{\beta\}\rangle = \exp\left(\sum_k (\alpha_k^*\,\beta_k - \tfrac{1}{2}|\alpha_k|^2 - \tfrac{1}{2}|\beta_k|^2)\right). \tag{104}$$

With the restriction (103), this sum is always convergent.

It is also easy to prove, as before, that a state which is initially a coherent state always remains coherent. In fact, if $|\psi, 0\rangle = |\{\alpha\}\rangle$, then $|\psi, t\rangle = |\{\alpha(t)\}\rangle$, where

$$\alpha_k(t) = \alpha_k \exp(-i\omega_k t). \tag{105}$$

Each $\alpha_k(t)$ follows the classical circular path, described with the appropriate constant angular velocity $-\omega_k$.

Using the eigenvalue equations (101) and (102), we can evaluate the expectation value of any normally-ordered product of the operators $\hat{a}_k$ and $\hat{a}_k^*$. For example, using the relations (85) and (86) connecting the electromagnetic fields to the generalized coordinates and momenta, we easily obtain

$$\langle\mathbf{B}(\mathbf{r}, t)\rangle = \langle\{\alpha(t)\}|\,\hat{\mathbf{B}}(\mathbf{r})|\{\alpha(t)\}\rangle = \sum_k \left(\frac{2\hbar}{\omega_k}\right)^{\frac{1}{2}} \nabla\wedge\mathbf{f}_k(\mathbf{r})\,\mathrm{Re}\,[\alpha_k \exp(-i\omega_k t)], \tag{106}$$

and

$$\langle\mathbf{E}(\mathbf{r}, t)\rangle = \langle\{\alpha(t)\}|\,\hat{\mathbf{E}}(\mathbf{r})|\{\alpha(t)\}\rangle = \sum_k -(2\hbar\omega_k)^{\frac{1}{2}}\,\mathbf{f}_k(\mathbf{r})\,\mathrm{Im}\,[\alpha_k \exp(-i\omega_k t)]. \tag{107}$$

These expectation values are of course solutions of Maxwell's equations. Correspondingly, for any solution of Maxwell's equation satisfying the chosen boundary conditions, we can find a coherent state in which the expectation values of the field operators are equal to the fields of this classical solution. The coherent state is the quantum state which best approximates the given classical field.

4.3 INTERACTION WITH A CLASSICAL CURRENT DISTRIBUTION

We can also show that the coherent states are the states produced by interaction with a classical current distribution, which plays the role of the forcing term in the harmonic oscillator considered earlier.

Let us suppose that within the volume under consideration there is an externally prescribed current distribution, $\mathbf{j}(\mathbf{r}, t)$, which we assume to vanish on the boundary. First, to find the appropriate Hamiltonian, we consider the purely classical problem. In the presence of this current distribution, the radiation-gauge vector potential satisfies eqn (49),

$$\varepsilon_0 \frac{\partial^2 \mathbf{A}}{\partial t^2} - \mu_0{}^{-1} \nabla^2 \mathbf{A} = \mathbf{j} - \varepsilon_0 \nabla \frac{\partial \phi}{\partial t} = \mathbf{j}_T. \tag{108}$$

Actually, the appropriate definition of $\mathbf{j}_T$ in this case is slightly different, because instead of requiring ϕ to vanish asymptotically as $r \to \infty$ (the condition which led to (48)), we should now require it to vanish on the boundary of our volume V. It is still true, however, that $\mathbf{j}_T$ is the divergenceless part of $\mathbf{j}$.

Now, if we insert the mode decomposition (83) for $\mathbf{A}$, then we may isolate the term corresponding to a single mode by taking the scalar product with the mode function $\mathbf{f}_k$ and integrating over V. Using the orthonormality relation (90), we obtain

$$\frac{\mathrm{d}^2 q_k}{\mathrm{d}t^2} + \omega_k{}^2 q_k = j_k, \tag{109}$$

where

$$j_k(t) = \int_V \mathrm{d}^3\mathbf{r}\ \mathbf{f}_k(\mathbf{r}) \,.\, \mathbf{j}(\mathbf{r}, t). \tag{110}$$

We have dropped the subscript T on $\mathbf{j}$ in (110), because the longitudinal part of $\mathbf{j}$ actually does not contribute. By using Green's theorem its contribution may be transformed into the sum of an integral involving $\nabla \,.\, \mathbf{f}_k = 0$ and a surface integral which vanishes because of the assumed boundary condition for ϕ.

The equation of motion (109) is that of a forced harmonic oscillator. It may be derived from the Hamiltonian function $H = H_F + H_1(t)$, where

$$H_1(t) = -\sum_k q_k j_k(t), \tag{111}$$

which may also be written in the form

$$H_1(t) = -\int_V \mathrm{d}^3\mathbf{r}\ \mathbf{A} \,.\, \mathbf{j}, \tag{112}$$

using (110) and (83).

We can now immediately write down the corresponding Hamiltonian for the quantized electromagnetic field interacting with a classical current. It is $\hat{H} = \hat{H}_F + \hat{H}_1(t)$, where

$$\hat{H}_1(t) = -\sum_k \hat{q}_k j_k(t) = -\int_V \mathrm{d}^3\mathbf{r}\, \hat{\mathbf{A}} \cdot \mathbf{j}. \tag{113}$$

Since this Hamiltonian is a sum of terms, each containing the variables corresponding to only a single mode, it affects each mode separately. Our previous analysis of the forced oscillator applies without modification to each individual mode. In particular, let us suppose as before that the external current distribution $\mathbf{j}(\mathbf{r}, t)$ is zero except during some finite time interval, $t_0 < t < t_1$, and that for $t < t_0$ the system is in its ground state, $|0\rangle$, or in other words that no photons are present before the current is switched on. Then at any time $t > t_1$, each mode will be in a coherent state, given by (38). Thus the system as a whole will be in a coherent state,

$$|\psi, t\rangle = |\{\alpha(t)\}\rangle \exp(i\phi), \tag{114}$$

where $\alpha_k(t) = \alpha_k \exp(-i\omega_k t)$, with

$$\alpha_k = \frac{i}{(2\hbar\omega_k)^{\frac{1}{2}}} \int_{t_0}^{t_1} \mathrm{d}t\, j_k(t) \exp(i\omega_k t) = \frac{i\tilde{j}_k(\omega_k)}{(2\hbar\omega_k)^{\frac{1}{2}}} \tag{115}$$

as in (39), and

$$\phi = \frac{1}{\hbar} \sum_k P \int_0^{+\infty} \frac{\mathrm{d}\omega}{2\pi} \frac{|\tilde{j}_k(\omega)|^2}{\omega_k^2 - \omega^2}, \tag{116}$$

as in (40).

It is easy to verify, using (106) and (107), that the expectation values of the fields in the state (114) are precisely the classical fields one would obtain by solving Maxwell's equations in the presence of the prescribed current distribution, with the given boundary conditions, and the initial condition that the fields vanish for $t < t_0$.

4.4 WAVE VECTOR REPRESENTATION

So far we have been considering an electromagnetic field confined in a cavity of finite volume V. However, we often want to consider a field which is not so confined. We can do this by considering the limit of infinite volume, but in so doing it is more convenient to start with a different set of boundary conditions.

Let us choose a cubic volume $V = L^3$, and impose periodic boundary conditions,

$$\phi(x+L, y, z) = \phi(x, y+L, z) = \phi(x, y, z+L) = \phi(x, y, z) \tag{117}$$

on each of the field components.

Now, since the differential equations (81) and the boundary conditions (82) satisfied by our mode functions were real, we could always choose these functions $\mathbf{f}_k$ to be real also. (The real and imaginary parts of any complex solution are themselves solutions.) The same is true with the boundary conditions (117). However, it is more convenient to choose the mode functions to represent plane waves. Each mode is labelled by a wave vector $\mathbf{k}$ and a polarization index $s = 1, 2$. To satisfy the periodicity conditions (117), we must restrict the allowable wave vectors by the conditions

$$k_x L = 2\pi n_x, \qquad k_y L = 2\pi n_y, \qquad k_z L = 2\pi n_z, \tag{118}$$

wher n_x, n_y, n_z are integers. The mode functions are chosen to be

$$\mathbf{f}_{\mathbf{k}, s}(\mathbf{r}) = \frac{1}{(\varepsilon_0 V)^{\frac{1}{2}}} \mathbf{e}_s(\mathbf{k}) \exp(i\mathbf{k} \cdot \mathbf{r}), \tag{119}$$

where $\mathbf{e}_1(\mathbf{k})$ and $\mathbf{e}_2(\mathbf{k})$ are mutually orthogonal unit vectors perpendicular to $\mathbf{k}$. These functions are normalized so that

$$\varepsilon_0 \int_V \mathrm{d}^3\mathbf{r}\, \mathbf{f}^*_{\mathbf{k}, s} \cdot \mathbf{f}_{\mathbf{k}', s'} = \delta_{\mathbf{k}, \mathbf{k}'} \delta_{s, s'}. \tag{120}$$

The use of complex mode functions necessitates some changes in our earlier formulae. In particular, in expansions like (83), (85), or (86), of real fields, the coefficients cannot any longer be taken to be real, and therefore in the quantum mechanical case cannot be identified with the Hermitian operators $\hat{q}_k$ or $\hat{p}_k$. Instead, we may separate the terms involving annihilation and creation operators, and write

$$\hat{\mathbf{A}}(\mathbf{r}) = \sum_{\mathbf{k}, s} \left(\frac{\hbar}{2\omega_k}\right)^{\frac{1}{2}} [\hat{a}_{\mathbf{k}, s}\, \mathbf{f}_{\mathbf{k}, s}(\mathbf{r}) + \hat{a}^*_{\mathbf{k}, s}\, \mathbf{f}^*_{\mathbf{k}, s}(\mathbf{r})], \tag{121}$$

and

$$\hat{\mathbf{E}}(\mathbf{r}) = \sum_{\mathbf{k}, s} \left(\frac{\hbar\omega_k}{2}\right)^{\frac{1}{2}} i[\hat{a}_{\mathbf{k}, s}\, \mathbf{f}_{\mathbf{k}, s}(\mathbf{r}) - \hat{a}^*_{\mathbf{k}, s}\, \mathbf{f}^*_{\mathbf{k}, s}(\mathbf{r})]. \tag{122}$$

It is clear that these expansions reduce to the earlier ones when the mode functions are real.

As before, the annihilation and creation operators satisfy commutation relations of the form

$$[\hat{a}_{\mathbf{k},s}, \hat{a}_{\mathbf{k}',s'}] = [\hat{a}^*_{\mathbf{k},s}, \hat{a}^*_{\mathbf{k}',s'}] = 0, \qquad [\hat{a}_{\mathbf{k},s}, \hat{a}^*_{\mathbf{k}',s'}] = \delta_{\mathbf{k},\mathbf{k}'}\delta_{s,s'}. \tag{123}$$

From these it is a matter of straightforward algebra to compute the commutators of the field operators. The commutator of any two field variables is given by a double sum over $\mathbf{k}, s$ and $\mathbf{k}', s'$, which is reduced to a single sum when we insert the relations (123). To perform the sum over s we use the fact that $\mathbf{e}_1(\mathbf{k})$ and $\mathbf{e}_2(\mathbf{k})$ span the plane normal to $\mathbf{k}$, so that

$$\sum_{s=1}^{2} e_{s,i}(\mathbf{k})\, e_{s,j}(\mathbf{k}) = \delta_{ij} - \frac{k_i k_j}{\mathbf{k}^2}. \tag{124}$$

(The right hand side is the projector on this plane.) The sum over $\mathbf{k}$ is of the form

$$\sum_{\mathbf{k}} \exp\,[i\mathbf{k}\,.\,(\mathbf{r}-\mathbf{r}')] = V\delta_3(\mathbf{r}-\mathbf{r}'),$$

where $\delta(\mathbf{r}) = \delta(x)\delta(y)\delta(z)$ is the three-dimensional Dirac delta function, or is obtained from this relation by differentiation. When we go through the algebra, we find that any two components of $\hat{\mathbf{E}}$ (or any two of $\hat{\mathbf{B}}$) commute, as do, for example, $\hat{E}_x$ and $\hat{B}_x$. A typical non-vanishing commutator is

$$[\hat{B}_x(\mathbf{r}), \hat{E}_y(\mathbf{r})] = \frac{i\hbar}{\varepsilon_0}\frac{\partial}{\partial z}\delta_3(\mathbf{r}-\mathbf{r}').$$

We learn therefore that although it is possible in principle to measure with arbitrary accuracy parallel components of $\mathbf{E}$ and $\mathbf{B}$, this is not the case for perpendicular components.

We are now in a position to consider the infinite-volume limit. We note that according to (118), the allowable wave vectors are distributed uniformly in $\mathbf{k}$ space, forming a cubic lattice with spacing $2\pi/L$. The volume of $\mathbf{k}$ space per point is thus $(2\pi/L)^3 = (2\pi)^3/V$. Thus in the limit as $L\to\infty$, the spacing becomes arbitrarily small, and the sum over $\mathbf{k}$ goes over to an integral,

$$\frac{(2\pi)^3}{V}\sum_{\mathbf{k}} \to \int \mathrm{d}^3\mathbf{k}. \tag{125}$$

Correspondingly, in this limit a Krönecker delta symbol goes over into a Dirac delta function,

$$\frac{V}{(2\pi)^3}\delta_{\mathbf{k},\mathbf{k}'} \to \delta_3(\mathbf{k}-\mathbf{k}'). \tag{126}$$

It is convenient to absorb a factor of $V^{\frac{1}{2}}$ into the definition of the annihilation and creation operators. We set

$$\hat{a}_s(\mathbf{k}) = V^{\frac{1}{2}} \hat{a}_{\mathbf{k},s}. \tag{127}$$

Then the commutation relation (123) becomes

$$[\hat{a}_s(\mathbf{k}), \hat{a}_{s'}{}^*(\mathbf{k}')] = V\delta_{\mathbf{k},\mathbf{k}'}\,\delta_{s,s'} \to (2\pi)^3\,\delta_3(\mathbf{k}-\mathbf{k}')\,\delta_{s,s'}, \tag{128}$$

by (126). Moreover, the decomposition (121) may be written, using (119), as

$$\hat{\mathbf{A}}(\mathbf{r}) = \frac{1}{V}\sum_{\mathbf{k},s}\left(\frac{\hbar}{2\varepsilon_0\,\omega_k}\right)^{\frac{1}{2}}[\hat{a}_s(\mathbf{k})\exp(i\mathbf{k}\,.\,\mathbf{r}) + \hat{a}_s{}^*(\mathbf{k})\exp(-i\mathbf{k}\,.\,\mathbf{r})]\,\mathbf{e}_s(\mathbf{k})$$

$$\to \int\frac{\mathrm{d}^3\,\mathbf{k}}{(2\pi)^3}\sum_s\left(\frac{\hbar}{2\varepsilon_0\,\omega_k}\right)^{\frac{1}{2}}[\hat{a}_s(\mathbf{k})\exp(i\mathbf{k}\,.\,\mathbf{r}) + \hat{a}_s{}^*(\mathbf{k})\exp(-i\mathbf{k}\,.\,\mathbf{r})]\,\mathbf{e}_s(\mathbf{k}). \tag{129}$$

by (125). This then is the correct mode decomposition for the infinite-volume case.

It should be remarked that at least two other normalization conventions are in common use. Many authors introduce an extra factor $(2\pi)^{-3/2}$ in (127), thereby eliminating the $(2\pi)^3$ from (128), and replacing it in (129) by $(2\pi)^{3/2}$. On the other hand, particle physicists who are interested in making formulae obviously invariant under relativistic Lorentz transformations tend to include an extra factor of $(2\omega_k)^{\frac{1}{2}}$.

All our previous formulae can be carried over with appropriate modifications to the infinite-volume case. Since the number of modes is no longer countable, it is impossible to label the states by the occupation numbers of the modes. However, we can still use the alternative labelling introduced in (96) and (97). We may define the one-photon state

$$|\mathbf{k}, s\rangle = \hat{a}_s{}^*(\mathbf{k})|0\rangle. \tag{130}$$

This state has a delta-function normalization,

$$\langle\mathbf{k}, s|\mathbf{k}', s'\rangle = (2\pi)^3\,\delta_3(\mathbf{k}-\mathbf{k}')\,\delta_{s,s'}. \tag{131}$$

An arbitrary one-photon state is a linear combination of states of this form,

$$|\psi\rangle = \sum_s\int\frac{\mathrm{d}^3\mathbf{k}}{(2\pi)^3}\,|\mathbf{k}, s\rangle\,\psi_s(\mathbf{k}).$$

It is normalized if

$$\sum_s\int\frac{\mathrm{d}^3\mathbf{k}}{(2\pi)^3}\,|\psi_s(\mathbf{k})|^2 = 1.$$

Similarly, the basic two-photon states are

$$|\mathbf{k}_1, s_1; \mathbf{k}_2, s_2\rangle = \hat{a}_{s_1}{}^*(\mathbf{k}_1)\hat{a}_{s_2}{}^*(\mathbf{k}_2)\,|0\rangle. \tag{132}$$

Their orthonormality relations are

$$\langle \mathbf{k}_1, s_1; \mathbf{k}_2, s_2|\mathbf{k}_1', s_1'; \mathbf{k}_2', s_2'\rangle = \langle \mathbf{k}_1, s_1|\mathbf{k}_1', s_1'\rangle \langle \mathbf{k}_2, s_2|\mathbf{k}_2', s_2'\rangle + \langle \mathbf{k}_1, s_1|\mathbf{k}_2', s_2'\rangle \langle \mathbf{k}_2, s_2|\mathbf{k}_1', s_1'\rangle.$$

The basic states (130) and (132) are eigenstates not only of the Hamiltonian

$$\hat{H}_F = \sum_s \int \frac{\mathrm{d}^3\mathbf{k}}{(2\pi)^3}\ \hbar\omega_k\, \hat{a}_s{}^*(\mathbf{k})\, \hat{a}_s(\mathbf{k}) \tag{133}$$

and the total photon number operator

$$\hat{N} = \sum_s \int \frac{\mathrm{d}^3\mathbf{k}}{(2\pi)^3}\ \hat{a}_s{}^*(\mathbf{k})\ \hat{a}_s(\mathbf{k}) \tag{134}$$

but also of any other linear function of the operators $\hat{a}_s{}^*(\mathbf{k})\hat{a}_s(\mathbf{k})$. In particular, we may define the total momentum operator,

$$\hat{\mathbf{P}} = \sum_s \int \frac{\mathrm{d}^3\,\mathbf{k}}{(2\pi)^3}\ \hbar\mathbf{k}\, \hat{a}_s{}^*(\mathbf{k})\ \hat{a}_s(\mathbf{k}). \tag{135}$$

Its eigenvalues in the states (130) and (132) are $\hbar\mathbf{k}$ and $\hbar(\mathbf{k}_1+\mathbf{k}_2)$, respectively.

A coherent state must now be labelled not by a sequence of complex numbers, but by a complex function $\alpha_s(\mathbf{k})$, defined so that

$$\hat{a}_s(\mathbf{k})|\{\alpha\}\rangle = \alpha_s(\mathbf{k})|\{\alpha\}\rangle. \tag{137}$$

To see how to expand it in terms of the states labelled by $\mathbf{k}$, s, we note first that for a single mode,

$$\hat{a}^{*n}|0\rangle = (n!)^{\frac{1}{2}}|n\rangle,$$

and hence that (100) may be written

$$|\{\alpha\}\rangle = \exp\left(\sum_k \alpha_k a_k{}^*\right)|0\rangle \exp\left(-\tfrac{1}{2}\sum_k |\alpha_k|^2\right).$$

Hence, going to the infinite-volume limit, we have

$$|\{\alpha\}\rangle = \exp\left(\sum_s \int \frac{\mathrm{d}^3\mathbf{k}}{(2\pi)^3}\ \alpha_s(\mathbf{k})\, a_s{}^*(\mathbf{k})\right)|0\rangle \exp\left(-\tfrac{1}{2}\sum_s \int \frac{\mathrm{d}^3\mathbf{k}}{(2\pi)^3}\,|\alpha_s(\mathbf{k})|^2\right). \tag{138}$$

Expansion of the first exponential would yield an expansion in terms of n-photon states. We note also that the scalar product of two coherent states is now

$$\langle\{\alpha\}|\{\beta\}\rangle = \exp\left(\sum_s \int \frac{d^3\mathbf{k}}{(2\pi)^3}\,[\alpha_s^*(\mathbf{k})\,\beta_s(\mathbf{k}) - \tfrac{1}{2}|\alpha_s(\mathbf{k})|^2 - \tfrac{1}{2}|\beta_s(\mathbf{k})|^2]\right) \tag{139}$$

5. Quantum Electrodynamics

We now turn finally to the fully quantized theory, in which the system to be studied includes both the electrons and the electromagnetic field. The Hamiltonian function for the corresponding classical system includes both the field energy (56), and the Hamiltonian function (60) for each electron. However, as we remarked earlier, we must be careful to avoid counting twice the Coulomb energy which is present in both. If we label the electrons by an index j, then the classical Hamiltonian may be written

$$H = \int d^3\mathbf{r}\,\tfrac{1}{2}(\varepsilon_0\,\mathbf{E}^2 + \mu_0^{-1}\,\mathbf{B}^2) + \sum_j \left\{\frac{1}{2m}[\mathbf{p}_j - e\mathbf{A}(\mathbf{r}_j)]^2 + V_j(\mathbf{r}_j)\right\}, \tag{140}$$

where $V_j(\mathbf{r}_j)$ is an external Coulomb potential, representing the effect of the atomic nuclei for the case of bound electrons. The Coulomb energy of the electrons themselves is contained in the first term, as the contribution of the longitudinal part of $\mathbf{E}$.

It is convenient to write the corresponding quantum mechanical Hamiltonian as a sum of terms in which the free parts (independent of e) are separated from the interaction parts. We write

$$\hat{H} = (\hat{H}_{0F} + \hat{H}_C) + \sum_j (\hat{H}_{0j} + \hat{H}_{1j} + \hat{H}_{2j}). \tag{141}$$

Here $\hat{H}_{0F}$, the free Hamiltonian of the electromagnetic field is the first term of (140) with the Coulomb energy removed, namely

$$\hat{H}_{0F} = \tfrac{1}{2}\int d^3\mathbf{r}\,(\varepsilon_0\,\hat{E}_T^{\,2} + \mu_0^{-1}\,\hat{B}^2) = \sum_k \hbar\omega_k\,\hat{a}_k^*\,\hat{a}_k. \tag{142}$$

In the present case, we are dealing with point particles, and thus the charge density is a sum of delta functions,

$$\rho(\mathbf{r}) = \sum_j e\delta(\mathbf{r} - \mathbf{r}_j).$$

Thus the Coulomb energy may be written

$$\hat{H}_C = \tfrac{1}{2}\sum_{j,j'}{}' \frac{e^2}{4\pi\varepsilon_0|\mathbf{r}_j - \mathbf{r}_{j'}|}. \tag{143}$$

Any term with $j = j'$ in this sum is clearly infinite. It represents the infinite Coulomb self-energy of a classical point particle. However, such a term is merely a constant addition to the Hamiltonian, which it is legitimate to exclude. The prime on the summation sign indicates that all terms with $j = j'$ are excluded. (There are in fact other contributions to the self-energy, corresponding to processes in which an electron emits and then reabsorbs a photon, which must also be subtracted, but we shall not be concerned with these.)

The free Hamiltonian for the jth electron (including the Coulomb potential of the nucleus if it is bound) is

$$\hat{H}_{0j} = \frac{1}{2m}\hat{\mathbf{p}}_j^2 + V_j(\hat{\mathbf{r}}_j). \tag{144}$$

The interaction parts are

$$\hat{H}_{1j} = -\frac{e}{m}\hat{\mathbf{p}}_j \cdot \hat{\mathbf{A}}(\hat{\mathbf{r}}_j) \tag{145}$$

and

$$\hat{H}_{2j} = \frac{e^2}{2m}\hat{\mathbf{A}}(\hat{\mathbf{r}}_j)^2, \tag{146}$$

where $\hat{\mathbf{A}}(\hat{\mathbf{r}}_j)$ is given in terms of photon annihilation and creation operators by (83) or (121), namely

$$\hat{\mathbf{A}}(\hat{\mathbf{r}}_j) = \sum_k \left(\frac{\hbar}{2\omega_k}\right)^{\frac{1}{2}} [\hat{a}_k \mathbf{f}_k(\hat{\mathbf{r}}_j) + \hat{a}_k^* \mathbf{f}_k^*(\hat{\mathbf{r}}_j)]. \tag{147}$$

We can now proceed to treat this Hamiltonian (142) by perturbation theory, taking as our free Hamiltonian

$$\hat{H}_0 = \hat{H}_{0F} + \sum_j \hat{H}_{0j}. \tag{148}$$

The eigenstates of $\hat{H}_0$ may be labelled by the occupation numbers n_k for each mode of the electromagnetic field, and by the quantum numbers n_j which label the energy levels of each electron. The various interaction terms will induce transitions between these eigenstates of $\hat{H}_0$. The term $\hat{H}_{1j}$ will induce transitions in which the state of the jth electron changes, while one photon is absorbed or emitted, while for $\hat{H}_{2j}$ two photons must be absorbed or emitted (or one absorbed and one emitted). The Coulomb term $\hat{H}_C$ will induce direct transitions involving two different electrons, with no accompanying change in the photon occupation numbers.

5.1 SPONTANEOUS EMISSION

As a first example, let us consider a system containing only a single electron, bound in an atom, and suppose that initially it is an eigenstate $|n\rangle$ of $\hat{H}_0$, with no photons present.

To first order in e, we need consider only the interaction term $\hat{H}_1$, not $\hat{H}_2$ (n or $\hat{H}_C$, which of course does not appear for the single-electron case). The lowest-order transition probability to a final state $|f\rangle$ is given by the analogue of (71) for a perturbation which contains no explicit time-dependence, namely

$$w_{fn} = \frac{2\pi}{\hbar} |\langle f|\hat{H}_1|n\rangle|^2 \, \delta(E_f - E_n). \tag{149}$$

Now, since the intial state contains no photons, the term in $\hat{H}_1$ containing a photon annihilation operator cannot contribute. We need only consider the second term in (147). The only final states for which there is a non-vanishing transition probability in lowest order are those containing a single photon, say in the kth mode, and with the electron in an eigenstate $|m\rangle$, that is, states of the same form $|f\rangle = |m; k\rangle$. From (145) and (147), the corresponding matrix element is

$$\langle m; k|\hat{H}_1|n\rangle = -\frac{e}{m} \sum_{k'} \left(\frac{\hbar}{2\omega_{k'}}\right)^{\frac{1}{2}} \langle m; k|\hat{\mathbf{p}} \cdot \mathbf{f}_{k'}{}^*(\hat{\mathbf{r}}) \hat{a}_{k'}{}^*|n\rangle.$$

The operator appearing in the matrix element is a product of an operator involving the variables describing the state of the electron, $\hat{\mathbf{r}}$ and $\hat{\mathbf{p}}$, and a photon creation operator, and the matrix element is therefore a product,

$$\begin{aligned}\langle m; k|\hat{\mathbf{p}} \cdot \mathbf{f}_{k'}{}^*(\hat{\mathbf{r}}) \, \hat{a}_{k'}{}^*|n\rangle &= \langle m|\hat{\mathbf{p}} \cdot \mathbf{f}_{k'}{}^*(\hat{\mathbf{r}})\,|n\rangle \, \langle k|\hat{a}_{k'}{}^*\,|0\rangle \\ &= \langle m|\hat{\mathbf{p}} \cdot \mathbf{f}_k{}^*(\hat{\mathbf{r}})\,|n\rangle \, \delta_{kk'}.\end{aligned}$$

Thus the only non-zero term in the sum over k' is that with $k' = k$, and we have

$$\langle m; k|\hat{H}_1|n\rangle = -\frac{e}{m}\left(\frac{\hbar}{2\omega_k}\right)^{\frac{1}{2}} \langle m|\hat{\mathbf{p}} \cdot \mathbf{f}_k{}^*(\hat{\mathbf{r}})\,|n\rangle.$$

In particular, if we use plane-wave mode functions (119) and assume, as in our earlier semiclassical calculation, that the dipole approximation is valid, then we obtain

$$\langle m; \mathbf{k}, s|\hat{H}_1|n\rangle = -\frac{e}{m}\left(\frac{\hbar}{2\omega_k}\right)^{\frac{1}{2}} \frac{1}{(\varepsilon_0 V)^{\frac{1}{2}}} \langle m|\hat{\mathbf{p}}|n\rangle \cdot \mathbf{e}_s^*(\mathbf{k})$$

$$= -ie\omega_{mn}\left(\frac{\hbar}{2\omega_k \varepsilon_0 V}\right)^{\frac{1}{2}} \langle m|\hat{\mathbf{r}}|n\rangle \cdot \mathbf{e}_s^*(\mathbf{k}). \tag{150}$$

What we are interested in physically is the probability of emission of a photon in a given direction within some angular range defined by a solid angle $\mathrm{d}\Omega$. To find this probability, we have to sum over a small range of values of the wave vector $\mathbf{k}$, using the relation (125) in the form

$$\frac{1}{V}\sum_{\mathbf{k}} \to \int \frac{\mathrm{d}^3\mathbf{k}}{(2\pi)^3} = \frac{1}{(2\pi)^3}\int k^2\,\mathrm{d}k\,\mathrm{d}\Omega = \frac{1}{(2\pi c)^3}\int \omega_k{}^2\,\mathrm{d}\omega_k\,\mathrm{d}\Omega. \tag{151}$$

Inserting (150) into (149), and performing this summation, we obtain for the probability per unit time of a transition from $|n\rangle$ to $|m\rangle$, with the emission of a photon in the angular range $\mathrm{d}\Omega$, the expression

$$w_{mn}(\mathrm{d}\Omega) = \frac{2\pi}{\hbar^2}\sum_{\mathbf{k}}\sum_{s} |\langle m; \mathbf{k}, s|\hat{H}_1|n\rangle|^2\,\delta(\omega_{mn}+\omega_k)$$

$$= \frac{\alpha\omega^3{}_{nm}}{2\pi c^2}\,\mathrm{d}\Omega \sum_{s} |\langle m|\hat{\mathbf{r}}|n\rangle \cdot \mathbf{e}_s^*(\mathbf{k})|^2, \tag{152}$$

using the definition (77) of α. The sum over polarizations may be performed using the formula (124).

If we are interested only in the total transition probability from $|n\rangle$ to $|m\rangle$, then we have to integrate over all directions. Since

$$\int \mathrm{d}\Omega\left(\delta_{ij} - \frac{k_i k_j}{\mathbf{k}^2}\right) = \frac{8\pi}{3}\delta_{ij},$$

the total transition probability per unit time is

$$w_{mn} = \frac{4}{3}\frac{\alpha\omega^3{}_{mn}}{c^2}|\langle m|\hat{\mathbf{r}}|n\rangle|^2. \tag{153}$$

5.2 INDUCED EMISSION AND ABSORPTION

It is actually unnecessary to recalculate the transition probabilities for induced emission and absorption processes, because we can make use of a general theorem which allows us to reduce the calculation (at least in lowest order)

to the earlier semiclassical one. However, we shall also consider later a different, independent calculation, to show that the results are indeed the same.

Let us suppose that the initial state contains a single electron, in the eigenstate $|n\rangle$ of $\hat{H}_0$, together with a coherent state of the electromagnetic field. This choice is motivated by the observation that a coherent state is the quantum state which best approximates a classical field. Now, the measurement of the transition probability for some atomic transition may be regarded as a measurement of some property of the state of the electromagnetic field. In other words, we are at liberty to regard our atomic system as a piece of measuring apparatus with which we can measure some observable, which must be some function of the photon annihilation and creation operators $\hat{a}_k$ and $\hat{a}_k{}^*$. It is not necessary for our purposes to know exactly what function this is. We now apply the multi-mode generalization of the result (32), namely

$$\langle\{\alpha\}|F(\hat{a}_k, \hat{a}_k{}^*)|\{\alpha\}\rangle = \langle 0|F(\hat{a}_k+\alpha_k, \hat{a}_k{}^*+\alpha_k{}^*)|0\rangle. \tag{154}$$

This shows that the expectation value in question is the same as the corresponding vacuum expectation value of an operator obtained from F by adding c-number terms to $\hat{a}_k$ and $\hat{a}_k{}^*$. On the other hand, a classical external electro-magnetic field would also be represented by just such extra c-number terms, added to these operators. Hence we may conclude that the probability of some atomic transition in the presence of an electromagnetic field which is initially in a coherent state $|\{\alpha\}\rangle$ is equal to the probability of the same transition in the presence of a classical external field, but with no photons present in the initial quantum state-vector. In lowest order this gives just the semiclassical result, although there will of course be higher-order correction terms corresponding to processes in which photons are emitted and perhaps reabsorbed by the atomic system.

To verify this theorem in a special case, let us compute the probability of absorption of a photon. We begin by assuming that the intial state contains N photons, all in a single mode k. Since we shall be considering only this mode, we drop the mode label k for the moment, and denote the state simply by $|n; N\rangle$.

To first order in e, we again need to consider only the interaction term $\hat{H}_1$. Because it is no more than linear in the photon annihilation and creation operators, it can have non-vanishing matrix elements only between states whose photon numbers differ by ± 1. These are

$$\langle m; N-1\,|\hat{H}_1|n; N\rangle = -\frac{e}{m}\left(\frac{\hbar}{2\omega_k}\right)^{\frac{1}{2}} N^{\frac{1}{2}}\,\langle m|\hat{\mathbf{p}}\,.\,\mathbf{f}_k(\hat{\mathbf{r}})\,|n\rangle \tag{155}$$

and

$$\langle m; N+1\,|\hat{H}_1|n; N\rangle = -\frac{e}{m}\left(\frac{\hbar}{2\omega_k}\right)^{\frac{1}{2}} (N+1)^{\frac{1}{2}}\,\langle m|\hat{\mathbf{p}}\,.\,\mathbf{f}_k{}^*(\hat{\mathbf{r}})\,|n\rangle.$$

This matrix element must be substituted into (149) to give the transition probability per unit time. The argument of the delta function, the difference in energy between the final and initial states, is $E_m \mp \hbar\omega_k - E_n$. Thus for an absorption process we obtain the result

$$w_{mn} = \frac{2\pi}{\hbar^2}\left(\frac{e}{m}\right)^2 \frac{\hbar}{2\omega_k} N\, |\langle m|\hat{\mathbf{p}} \cdot \mathbf{f}_k(\hat{\mathbf{r}})|n\rangle|^2\, \delta(\omega_{mn} - \omega_k),$$

or, in the dipole approximation

$$w_{mn} = (2\pi)^2 \frac{\alpha N \omega_k c}{V} |\langle m|\hat{\mathbf{r}}|n\rangle \cdot \mathbf{e}_s(\mathbf{k})|^2\, \delta(\omega_{mn} - \omega_k). \tag{156}$$

It is easily seen that it makes no real difference if we replace the state with definite photon number by a coherent state. A coherent state is a linear superposition of states with different values of N, but since each N leads to a final state with a different photon number, their contributions to the transition probability add incoherently. The net effect is simply to replace N in (156) by the expectation value of $\hat{N}$ in the coherent state. Since the intensity in an N-photon state is the photon number density N/V multiplied by the energy of each photon $\hbar\omega_k$, and by the velocity c, $I = N\hbar\omega_k c/V$, the result (156) agrees precisely with the semiclassical result (75).

We can also consider initial states which contain photons in several different modes. If we choose a state containing n_k photons in the kth mode, then we see that the absorption of a photon from one mode leads to a state orthogonal to that obtained by absorption from a different mode. Thus, again, these contributions to the transition probability add incoherently. The resulting expression, obtained by summing over a range of values of $\mathbf{k}$ agrees with the semiclassical result (76).

5.3 SCATTERING PROBLEMS

As an example of a rather different kind, let us consider the scattering of radiation by an electron in an atom. We suppose that the electron is initially in its ground state, $|g\rangle$, say, and that there is a single photon present in the kth mode. Thus the initial state may be denoted by $|g; k\rangle$. We seek to calculate the probability of a transition to a final state in which there is again a single photon present, but in a different mode, and in which the electron is left in a state $|m\rangle$ (which may once more be the ground state, or may be one of the excited states, or indeed an unbound state, if the photon is sufficiently energetic to ionize the atom). The final state in which we are interested is therefore $|m; k'\rangle$.

Now, since $\hat{H}_1$ is linear in the photon annihilation and creation operators, it cannot in first order induce transitions in which the photon numbers of

two different modes change. Thus there is no matrix element to order e, and we have to go to order e^2 to obtain a non-vanishing contribution. In that order, there are two distinct contributions. In first-order perturbation theory, there is a contribution from the term $\hat{H}_2$ in the Hamiltonian, which is of order e^2, and, moreover, $\hat{H}_1$ itself can contribute in the second-order of perturbation theory.

Let us consider first the effect of $\hat{H}_2$. Since we have to remove a photon from the kth mode, and create a photon in the k'th, the relevant term is the one proportional to $\hat{a}_{k'}{}^*\hat{a}_k$. Identifying the coefficient of this term from (146) and (147), we find that its contribution is

$$\langle m; k'|\hat{H}_2|g; k\rangle = \frac{e^2}{m}\frac{\hbar}{2(\omega_k\omega_{k'})^{\frac{1}{2}}}\langle m|\mathbf{f}_{k'}{}^*(\hat{\mathbf{r}})\,.\,\mathbf{f}_k(\hat{\mathbf{r}})|g\rangle, \tag{157}$$

or, if we specialize to the case of plane-wave modes,

$$\langle m; \mathbf{k}', s'|\hat{H}_2|g; \mathbf{k}, s\rangle = \frac{e^2\hbar}{2m\varepsilon_0 V(\omega_k\omega_{k'})^{\frac{1}{2}}}\langle m|\exp[i(\mathbf{k}-\mathbf{k}')\,.\,\hat{\mathbf{r}}]|g\rangle\,\mathbf{e}_{s'}{}^*(\mathbf{k}'),\mathbf{e}_s(\mathbf{k}). \tag{158}$$

If the frequency of the photon is sufficiently low to make the dipole approximation valid, then we obtain a non-vanishing matrix element only in the special case $m = g$, so that the scattering is necessarily elastic. It is clear moreover, that in this case the matrix element would be the same for any initial state. This is what we should expect physically. In the long wavelength limit, the details of the atomic structure are irrelevant, and the photon scatters as if from a free electron.

Let us for the moment ignore the contribution of $\hat{H}_1$, and suppose that (158) represents the complete matrix element. Then we may substitute it into (149) to find the transition probability per unit time. The argument of the energy-conserving delta function is of course $E_m+\hbar\omega_{k'}-E_g-\hbar\omega_k = \hbar(\omega_{mg}+\omega_{k'}-\omega_k)$. To find the probability that the final-state photon emerges in a direction within the solid angle $\mathrm{d}\Omega$, we may sum over a range of values of $\mathbf{k}'$, as in (151). Now, the flux of photons in the initial state is the photon number density, $1/V$, multiplied by the velocity c, say $f = c/V$. Dividing the transition probability per unit time by this flux, we obtain the differential cross section $\mathrm{d}\sigma$ for this process. It is given explicitly by

$$\mathrm{d}\sigma = \frac{V}{c}\frac{V}{(2\pi c)^3}\int \omega_{k'}{}^2\,\mathrm{d}\omega_{k'}\,\mathrm{d}\Omega\,\frac{2\pi}{\hbar^2}\left|\frac{e^2\hbar}{2m\varepsilon_0 V(\omega_k\omega_{k'})^{\frac{1}{2}}}\langle m|\exp[i(\mathbf{k}-\mathbf{k}')\,.\,\hat{\mathbf{r}}]|g\rangle\right.$$
$$\left.\times\,\mathbf{e}_{s'}{}^*(\mathbf{k}')\,.\,\mathbf{e}_s(\mathbf{k})\right|^2\delta(\omega_{mg}+\omega_{k'}-\omega)$$

or

$$d\sigma_{mg} = \frac{\omega_{k'}}{\omega_k} r_0^2 d\Omega \left| \langle m| \exp\left[i(\mathbf{k}-\mathbf{k}')\cdot\hat{\mathbf{r}}\right] |g\rangle \right|^2 |\mathbf{e}_{s'}^*(\mathbf{k}')\cdot\mathbf{e}_s(\mathbf{k})|^2, \tag{159}$$

where we have introduced the classical electron radius

$$r_0 = \frac{\alpha\hbar}{mc} = \frac{e^2}{4\pi\varepsilon_0 mc^2}. \tag{160}$$

In (159), the magnitude of $\mathbf{k}'$ is fixed by the energy-conservation condition $\omega_{k'} = \omega_k - \omega_{mg}$.

If the frequency of the incident photon is sufficiently low, then the dipole approximation will be applicable. Then the only non-vanishing cross section is that for elastic scattering,

$$d\sigma_{gg} = r_0^2 d\Omega\, |\mathbf{e}_{s'}^*(\mathbf{k}')\cdot\mathbf{e}_s(\mathbf{k})|^2. \tag{161}$$

This is the classical Thomson scattering cross section, which can also be obtained by either classical or semiclassical methods. The total scattering cross section, obtained by integrating over all angles, and summing over s', is

$$\sigma = \frac{8\pi}{3} r_0^2.$$

For higher frequencies, the matrix element in (159) must be evaluated explicitly. This formula can also be used to calculate the ionization cross section, by summing over all unbound states $|m\rangle$ of the electron, up to the maximum available energy.

An exactly similar calculation can be performed for free electrons, leading to an expression identical in form to (159). In that case, we may label the electron states by the momentum $\mathbf{p}$. If we choose the electron to be initially at rest ($\mathbf{p} = \mathbf{0}$), and finally in the momentum eigenstate $|\mathbf{p}'\rangle$, then we obtain a non-vanishing matrix element only when the momentum-conservation condition $\mathbf{p}' + \hbar\mathbf{k}' = \hbar\mathbf{k}$ is satisfied. When combined with the energy-conservation condition $E_{\mathbf{p}'} + \hbar\omega_{k'} = \hbar\omega_k$, this condition yields the formula for the frequency shift in Compton scattering.

So far, we have ignored the contribution of $\hat{H}_1$. To evaluate it, we need second-order perturbation theory. By substituting the first-order solution $u_m^{(1)}$, given by (68), into (67), we obtain $u_m^{(2)}$. Then from this we may evaluate the second-order transition probability per unit time. We shall not go through the algebra in detail, because it involves certain additional complications which would take us too far afield, but merely quote the result. The required

modification of the first-order formula is obtained by replacing the matrix element $\langle m|\hat{H}_1|i\rangle$ from an initial state $|i\rangle$ to a final state $|m\rangle$ by

$$\langle m|\hat{H}_2|i\rangle - \sum_n \frac{\langle m|\hat{H}_1|n\rangle\langle n|\hat{H}_1|i\rangle}{E_n - E_i}, \tag{162}$$

where the sum is over the complete set of eigenstates $|n\rangle$ of $\hat{H}_0$.

In the present case, the first-order term is zero (except for the contribution of $\hat{H}_2$ considered above). Since $\hat{H}_1$ can only create or annihilate photons one at a time, one factor of $\hat{H}_1$ must annihilate the initial-state photon, and the other must create the final-state photon. These processes may occur in either order. Thus the intermediate states which contribute to the sum are either states $|n\rangle$ with no photons, or states $|n; k, k'\rangle$ containing two photons. (Here in contrast to (162), n labels the states of the atom alone.) Inserting the expressions for the matrix elements of $\hat{H}_1$, we see that the term which must be added to the matrix element (157) of $\hat{H}_2$ is

$$-\frac{e^2}{m}\frac{\hbar}{2(\omega_k\omega_{k'})^{\frac{1}{2}}}\sum_n \frac{\langle m|\hat{\mathbf{p}}\cdot\mathbf{f}_{k'}^*(\hat{\mathbf{r}})|n\rangle\langle n|\hat{\mathbf{p}}\cdot\mathbf{f}_k(\hat{\mathbf{r}})|g\rangle}{E_n - E_g - \hbar\omega_k}$$

$$+ \frac{\langle m|\hat{\mathbf{p}}\cdot\mathbf{f}_k(\hat{\mathbf{r}})|n\rangle\langle n|\hat{\mathbf{p}}\cdot\mathbf{f}_{k'}^*(\hat{\mathbf{r}})|g\rangle}{E_n - E_g + \hbar\omega_{k'}}. \tag{163}$$

A particular term in the sum over n will yield a large contribution when the incident photon frequency is close to ω_{ng}. Each energy level gives rise to a resonance of this kind in the scattering. (Higher-order terms in the perturbation series have the effect of giving each level a finite width, thus removing the infinity in (163).) In general, the denominator of the second term in the brackets can never be small, so that this term is negligible.

Terms like (163) should also be included in the case of a free electron. However, if the electron is initially at rest, their contribution is identically zero. This is because of the factor $\hat{\mathbf{p}}$ in the matrix elements, and the fact that the initial state $|g\rangle$ is an eigenstate of $\hat{\mathbf{p}}$ with eigenvalue $\mathbf{0}$. Thus for a free electron initially at rest the cross section is given simply by (159).

Concluding Remarks

The theory we have been discussing is still quite far from being the full theory of quantum electrodynamics, because it involves several approximations which, though legitimate for optical frequencies, must be discarded when we come to consider higher frequencies.

We have throughout treated the electrons as spinless, and ignored the

interaction between the electromagnetic field and the magnetic moment of the electron. This approximation is not essential, and could fairly easily be removed. We should then have to consider a system with an extra term, of the form (constant) $\times \hat{\boldsymbol{\sigma}} \cdot \hat{\mathbf{B}}$, in the Hamiltonian.

A much more important approximation is that we have treated the electrons non-relativistically. The transition to a relativistic theory involves a lot more than merely a change in the relationship between energy and momentum. It also requires that we allow for the possibility of pair creation and pair annihilation. A high-energy photon striking an atom may not merely scatter but produce one or more pairs, consisting of an electron and a positron. This possibility also leads to the phenomenon of vacuum polarization, the creation and subsequent reannihilation of a virtual pair. Because of this effect, the observed charge of the electron, e, is not the same as the constant e_0 which appears in the Hamiltonian. It is modified because, even in a vacuum, an electron is in a polarizable medium. Perturbation theory applied to quantum electrodynamics tends to give infinite results, beyond the lowest order. However, it has been shown that all the infinities may be regarded as contributions to just two constants, relating the observed charge and mass of the electron, e and m, to the constants which appear in the Hamiltonian, e_0 and m_0. Expressed in terms of e and m, rather than e_0 and m_0, observable quantities such as scattering probabilities are always finite. The predictions of the theory, in particular for the anomalous magnetic moment of the electron, and the Lamb shift in hydrogen-like atoms, are among the best verified predictions of any existing physical theory.

In practical terms, if we are dealing with radiation of optical frequencies (and even more, of course, for microwaves), then relativistic and spin corrections are generally small. They would have to be included if we wanted results of great accuracy, so that there would be little sense in going to high orders of perturbation theory without including them, but if we are satisfied with the leading order, there is no need to do so. It is also true that for many problems the quantization of the radiation field is unnecessary. As we have seen, processes of induced emission and absorption of radiation can be handled semiclassically, with the same results (in lowest order) as those provided by the full quantum mechanical treatment. Even the process of spontaneous emission can, in a sense, be reproduced in a semiclassical theory. We can take the expectation value of the electric current operator, and treat that as a source of the field, inserting it as an inhomogeneous term in Maxwell's equations. Then we do not get precisely the results of the quantum theory, but something approximating to them. In this semiclassical theory, the excited states of an atom would, strictly speaking, be stable because the expectation value of $e\hat{\mathbf{p}}/m$ is zero in any such state. However, if the initial state is not precisely an eigenstate of energy, but contains a small component of the ground state, then the

expectation value is non-zero, so that the system does radiate, and will decay eventually to its ground state, though not initially according to an exponential law. The fact that the emitted radiation obeys Planck's famous relationship between energy and frequency is a consequence of the quantization of the atomic energy levels and does not, of itself, require the quantization of the radiation field. Field quantization is, however, needed to explain such things as photon-counting statistics.

Quantum Theory of Coherence†

R. J. GLAUBER

1. INTRODUCTION

Quantum electrodynamics is a fairly mature field these days; it is about forty-two years old. The curious thing is that although it has become an immensely accomplished and versatile study as it has developed from optics, there has been very little connection maintained between quantum electrodynamics and optics. As you know, the two studies overlap completely in principle, but until a few years ago the connection between them remained only rudimentary. We shall consider first the question of why that situation existed for so long. The much closer liaison which we have now between these two fields developed at approximately the same time as the laser appeared on the scene, and one of its by-products has been a particularly useful way of describing laser beams. The primary impetus to develop the theory, however, lay in a number of interesting questions about the behaviour of photons which dated from earlier years: we shall see below what those questions were.

Perhaps the easiest way to obtain an overall picture of quantum electrodynamics is to think of the electromagnetic spectrum. Just how far we say the spectrum extends toward its low-frequency end is more or less a matter of taste. For example, it could be maintained that we have seen fields oscillating at something like 10 Hz or thereabouts; on the other hand, at the violet end of the spectrum, the quanta become more and more energetic as we go through the regions of X-rays, and γ-rays, etc. The γ-ray spectrum is rather extensive and it is not unlikely that we have seen the effects of γ-ray quanta whose frequencies are of the order of 10^{27} Hz. So we have something like 26 powers of 10 covering the spectrum with the visible band of the spectrum more or less in the middle as we observe it. The vast range of phenomena that radiations of these different frequencies produce, of course, recapitulates the entire history of physics.

Probably the greatest of the accomplishments of present day physics lies in the fact that one single theory explains everything that occurs over the vast

† These notes have been prepared with a minimum of editing, directly from tape recordings of Professor Glauber's lectures

range of the electromagnetic spectrum. Exactly the same things are happening, from a mathematical standpoint, in the radio frequency range and in the highest frequency range of the γ-rays produced only in cosmic radiation. It is all one theory; it has to be one theory because of the character of the Lorentz transformation, because we know that quanta of one frequency can be turned into quanta of any other frequency simply by observing them from a moving system. While that does not sound like a very practical thing for people to do (turning radio waves into γ-rays, let us say), it is a very practical thing for electrons; electrons are easily accelerated to the appropriate velocities. In fact one way we have these days of creating a monochromatic γ-ray beam with energies of hundreds of millions of volts is to let ordinary visible quanta collide with electron beams in a linear accelerator. That has been done at Stanford and it does provide a usable γ-ray beam. The electrons thus "see" ordinary light quanta as energetic γ-rays and in colliding with them transfer just a little momentum to them, but that little momentum looks like an awful lot in the laboratory frame, and, indeed, those visible quanta come out at very substantial energies. That ought to be evidence enough that the spectrum is all one unit, and it must be one theory which embraces the whole.

The different physical qualities which characterize the different ends of the spectrum are completely at variance with one another. From a quantum mechanical standpoint, in the low frequency end of the spectrum we have fantastic quantum densities. If we asked, "How many quanta are there, in, for example, a cubic wavelength of radio broadcast radiation?" (of course, the wavelength gets a little large, for that kind), the answer tends to be an astronomical number. It is not unusual to see 10^{30}–10^{40} quanta per cubic wavelength. We have, of course, very efficient sources of such radiation, ones which produce vast numbers of essentially identical quanta. So at the lower frequency end of the spectrum we have lots of feeble-energy quanta. At the other end of the spectrum we have extremely energetic quanta; individual ones can do simply catastrophic things to atomic or nuclear systems. We have no trouble at all detecting such quanta by almost macroscopic means, but we have very very few quanta there at all. Now, in a sense we have still fewer quanta if we compare the number of quanta with how many we could have. The idea of mode density in the field is considered elsewhere in this chapter. It is slightly artificial for optics to consider the field within a finite volume, but it is very convenient to do so. It is convenient because it turns the continuum of oscillation of the field into a discrete set of modes which can be counted. It is a little artificial because we scarcely know what a perfect reflector is for, say, γ-rays at 100 MeV. But let us imagine such a thing just for the sake of defining the modes of the field mathematically. If we do that, the density of the modes per unit frequency interval rises as the square of the frequency and so the modes become enormously dense at very high frequencies.

But as the frequency increases the number of quanta produced by all reasonable sources goes down dramatically. Since nearly all the modes are thus empty there is a sense in which all high frequency quantum beams are exceedingly weak. We are familiar, on the other hand, with quantum beams of enormous intensity at the low frequency end of the spectrum where we are dealing with quantum energies which are exceedingly small compared even to thermal excitation energies. We have to go up to frequencies corresponding to wavelengths of 0·1 mm, or thereabouts, before we get to quantum energies which are comparable even with rather low thermal energies. So, quantum phenomena are masked by thermal noise and are not visible in the low frequency end of the spectrum which thus remains the natural domain of classical theory. We should have a rather hard time indeed persuading a radio frequency engineer that there is any need to deal in quantum mechanical terms with the output of a broadcast station. On the other hand, we could not be more familiar with the use of quantum theory in dealing with the high frequency region, essentially infra-red frequencies and higher, and that has always been the domain of quantum mechanics.

There are two other qualities of these two regions which are worth mentioning. One is the degree of control we have of the radiation. Our knowledge of classical radiation and how to control it in macroscopic terms is about 100 years old or so and we have achieved a great degree of versatility in being able to control radiation of relatively low frequencies. We can make the fields in oscillators do just about anything that the Maxwell equations will allow them to do, as a function both of the space and time variables. But, what happens, crudely speaking, as we proceed to higher frequencies is that we lose more and more control over what the field does. At low frequencies we have temporal control; we are able to produce, for example, quite monochromatic oscillations with steady amplitudes. As the frequency rises we sacrifice that possibility rather quickly. We still have monochromatic radiations available to us in the visible region, but only because of the accident of nature that atoms happen to have very well-defined frequencies of oscillation. As we go on into the X-ray region there is more and more radiation that just comes from the continuum of atomic excitations. Beyond the energies of a few million volts, we have even gone past the discrete levels of the nuclei and we then deal only with continuum radiation. We have virtually no monochromatic sources in the extremely high frequency region, except, perhaps the strange one I mentioned which involves the Compton scattering of monochromatic visible photons at a particular angle. That is just about the only monochromatic source that has been proposed in the multi-million volt region. In other words, we lose control of the time dependence of the fields as we increase the frequency. We also lose, somewhat more slowly, but none-the-less positively, control of the spatial behaviour of the fields. Optics in the visible region is a

field in which we are able to maintain very accurate control of the spatial behaviour of the field, for example, in the creation of interference patterns and the like. But it is quite difficult at the same time to maintain very accurate control over the time dependence of optical fields. Even though optical fields may be fairly monochromatic their amplitudes tend to fluctuate uncontrollably. We have thus to ask: "How does the field behave? What does the indefiniteness of the frequency of an optical field mean? What kind of time variations of the field are responsible for the line-broadening?" We shall see something very specific about that presently. What we shall find is that all radiation which does not come from specifically stabilized sources, i.e. all natural radiation is, in a fundamental sense, noise. It may have any sort of spectrum but it is what the electrical engineer would simply call coloured noise. The whole science of optics, in a sense, has just consisted of the constructive use of noise.

Quantum phenomena are easily observed in the visible region. They are observed in the sense that a photon with an energy of a few volts is perfectly detectable in a photomultiplier. Not very much work, historically, was done by detecting individual quanta in the visible region; the technique was thought of mainly as a way of detecting rather weak light beams. It was thought of, in other words, only as a means of intensity measurement and there are other and simpler ways of measuring low intensities, for example by integrating over a long period of time using photographic detection. There was, in all, remarkably little interest in quantum detection. That has changed a good deal within the last few years with the entry of a certain amount of technology into the picture—technology and a few very clever ideas.

Historically, the first development based on quantum detection was in an experiment by Hanbury-Brown and Twiss in 1956.[4] (To be perfectly precise, it was not quantum counting in the original experiment, it was the examination of fluctuations in the photo-current, but in all the subsequent experiments it has literally been quantum counting.) In counting quanta we can detect something more than just the intensity of the field; we can detect correlations within the field, and out of that fact has grown a whole idea of statistical studies of photons in the field. The techniques of measurement are ones which are carried down from the high frequency end of the spectrum. They are the techniques of high energy physics, a gift, if you like, from the people who have preoccupied themselves with that end of the spectrum. And there is one more great development, which is the development of the laser itself.

The laser radiates in approximately the same way as the classical radiator to which we have been accustomed for a long time at the low frequency end of the spectrum. That is done by exploiting atomic transitions and macroscopic techniques as well. What results, in effect, is a transfer of the ideas and the techniques of the low frequency end of the spectrum upwards in frequency

by a factor of perhaps 10^5 or so. The result is that now there is a region of the spectrum extending over the visible frequencies and into the near ultra violet in which we can both generate quanta coherently and detect them quantum mechanically, i.e. with devices which detect the individual photons. Now, lasers are extremely intense sources of essentially identical photons. Such sources were previously available only at radio frequencies but, at optical frequencies, the photons were so sparse that we never had to think of two identical photons as actually being in the field at the same time. That, crudely speaking, is still the case at X-ray frequencies and beyond. There are so many modes available to distribute the photons into, that we simply never have, to any noticeable degree, two photons occupying a single mode of the field at the same time, and so the statistical properties of the fields at X-ray frequencies and beyond are in a deep sense rather trivial—nothing very interesting goes on.

The laser produces almost arbitrarily intense photon beams in the visible region of the spectrum and experiments with these beams can lead to all sorts of quantum populations of the modes of the electromagnetic field. We can use quantum detection to investigate the statistical properties of these populations. That is, in essence, the change that has taken place. It has been necessary, therefore, for theory to respond in a way which permits us to deal fairly easily with many-photon problems. The development of the theory, until a few years ago, really went no further than the use of classical theory in the low frequency domain and at high frequencies quantum mechanics in its simplest form, dealing with one or two photons at one time. There are many reasons why the classical limit of quantum electrodynamics was never very fully developed, even though we had every reason to be confident that quantum electrodynamics has the correct classical limit. The most fundamental of these reasons is the smallness of the number $e^2/(4\varepsilon_0 hc)$. Its smallness means that we can proceed with great accuracy in doing calculations with individual charged particles by a perturbation expansion. In point of fact, it has been very difficult in the past in quantum electrodynamics (q.e.d.) to do anything other than this sort of approximation, but the perturbation expansion becomes vastly more difficult as you proceed from one order to the next. The tendency, therefore, has been to restrict our attention in q.e.d. to problems involving very few quanta at a time, and that is frequently a good approximation. So, if we are dealing with individual electrons, as we often are in this region of the spectrum, it is quite sufficient to say that we have an electron with only one or two photons in the field. If we are terribly ambitious and work for half a lifetime we might do a problem with three or four photons in the field, but scarcely more. There was therefore no great enthusiasm, nor was there a great need for dealing quantum mechanically with problems in which we had vast numbers of photons in the field at the same time. These days, however, we have very little alternative to doing just that in the visible region for the reasons

explained above, and that simply means that we must develop other approximations. The approximations cannot be precisely the approximations of familiar perturbation theory. We have no choice but to deal with very considerable numbers of photons at once.

The sort of optics which existed until recently (an immensely successful study) has been largely constructed in classical terms. The description of interference experiments, diffraction experiments, and the like, has usually been constructed in classical terms that were observed to work, and the advent of the quantum theory really lent a certain support to those rather *ad hoc* procedures of calculation in the following sense. It is very easy to calculate diffraction patterns in the one-photon approximation. We say the field has one photon in it and ask for the probability that one photon is seen in a particular position on the screen. Then we have to interpret that in statistical terms; we have to imagine that the one-photon experiment has been done time and again and ask where, statistically, that photon turns out to be. The average density of the photon, the statistical density, then agrees with the classical diffraction pattern and there can be no great surprise about that. Calculating the expectation value of the field intensity in single-photon states can be reduced in fact to much the same mathematical procedure as the classical intensity calculation. All of the calculations that we could do in single photon terms were quickly found thus to agree with the results of the classical calculations and to lend a good deal of support to the notion that the classical procedures were correct.

The kind of experiment we have described gives us a good way of characterising the older optics, the optics prior to the developments of the past few years. In all of these traditional optical experiments we imagine a limiting process in which the field intensity is allowed to go to zero, but the detection time is allowed to become infinite—in other words, the experiment of Taylor[1] described by Sillitto in his introductory remarks in this volume. All of the traditional optical experiments (perhaps that is a good way of defining tradition) have the quality that we could go to zero intensity and just increase the detection efficiency and we would detect basically the same thing, the same diffraction pattern. Those are one-photon experiments. They are the type of experiment we can describe by saying there is only one photon in the field at a time. But that description does not hold, for example, for the Hanbury-Brown–Twiss type of experiment which detects correlations between pairs of photons.

The Hanbury-Brown–Twiss experiment is really the first one in the higher frequency range which stays completely outside that tradition. Since that experiment, there have been a great many which step outside the tradition and which cannot be dealt with at all in terms of one-photon optics. To begin with, we can understand in such terms essentially nothing of the functioning

of the laser. The laser is intrinsically a non-linear device; it only works when the field intensities become so high that the photons know a great deal about each other's presence. That could not be less typical of one-photon optics. More generally, the availability of laser beams has made it possible to develop a new area of optics, non-linear optics. In non-linear optics, we do dramatic things such as sewing pairs of photons together to make new photons of twice the frequency. Needless to say, we learn nothing of phenomena like that from one-photon optics, and, as a matter of fact, a great deal in the phenomenon of harmonic generation depends upon the tendency of photon packets to overlap, or not to overlap, in any given beam. What we are beginning to suggest is that the description of the electromagnetic field which was used in the traditional optics is really inadequate. While we can describe the one-photon experiments by means of a very small group of parameters, in general, or a very restricted class of correlation functions, to characterise an electromagnetic field more fully, and, in particular, to do it in quantum mechanical terms, we need to know a great deal more.

2. Classical Theory

What we shall do now is to develop something of the newer and fuller description of the field that we need in order to deal with this much broader class of phenomena. Let us do this by first making contact with the classical noise theory and let me introduce the notation for the mode functions. This is material that Kibble went over (pp. 11–52, this volume). Imagine that we are dealing with the field inside an enclosed volume. The specification of the boundary conditions of that volume is really not essential to us but we need some sort of boundary condition in order to complete the specification of the problem. The boundary conditions could be those of a perfectly conducting surface; that, however, condemns us to talking about standing waves and makes it difficult to deal with progressive waves if we are really dealing with a problem in free space in which the very idea of boundaries is unnatural. A simpler thing to do is to use periodic boundary conditions. (All we need agree upon here is the existence of the mode functions; the boundary conditions are really immaterial, that is why they are left unspecified.)

We consider, then, the field inside a closed volume and, for an appropriate set of frequencies ω_k, we introduce a set of vector mode functions $\mathbf{u}_k(\mathbf{r})$ with mode index k which satisfy the wave equation

$$\left(\nabla^2 + \frac{\omega_k^2}{c^2}\right) \mathbf{u}_k(\mathbf{r}) = 0, \tag{1}$$

the transversality condition

$$\nabla \cdot \mathbf{u}_k(\mathbf{r}) = 0, \tag{2}$$

and the boundary conditions on the electric fields. The set of mode functions are orthogonal if the modes are of different frequency, or they can be made orthogonal if they are part of a degenerate subspace.

The orthonormality condition is

$$\int \mathbf{u}_k^*(\mathbf{r}) \cdot \mathbf{u}_l(\mathbf{r})\, d\mathbf{r} = \delta_{kl}. \tag{3}$$

The electric field may be broken into two terms; we shall call them the positive and negative frequency parts and define them by convention thus

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}^{(+)}(\mathbf{r}, t) + \mathbf{E}^{(-)}(\mathbf{r}, t), \tag{4}$$

where $\mathbf{E}^{(+)}$ is the sum of all terms which vary as $\exp(-i\omega_k t)$ for all $\omega_k > 0$. All the positive frequency terms are in $\mathbf{E}^{(+)}$ and all the complex conjugate terms are in $\mathbf{E}^{(-)}$. Therefore, we have

$$\mathbf{E}^{(-)} = (\mathbf{E}^{(+)})^*. \tag{5}$$

These are classical fields, we are not dealing with operators yet.

Now we expand a component of $\mathbf{E}^{(+)}$ in terms of the set of mode functions $\mathbf{u}_k$ and obtain

$$\mathbf{E}^{(+)}(\mathbf{r}, t) = \sum_k \mathscr{C}_k \mathbf{u}_k(\mathbf{r}) \exp(-i\omega_k t). \tag{6}$$

If there is no source connected to the field, these Fourier coefficients $\mathscr{C}_k$ are constants of motion. We shall deal with a field remote from its source; let us say the field is radiated from a distant nebula; we can regard it, to an excellent approximation, as being source-free. The field is then described completely by this set of coefficients (complex numbers) $\mathscr{C}_k$. The question is, "What are they?" If the field is perfectly well defined, if there is no randomness in it whatever, then we know, in principle, precisely what those numbers $\mathscr{C}_k$ are. To a communications engineer, a noiseless field would be one for which we know all the complex numbers, $\mathscr{C}$. In practice, there is no such thing as a completely noiseless field, we do not know the numbers $\mathscr{C}_k$, and the most accurate specification of a classical field, in statistical terms, can only be through a specification of the probability distribution of these complex numbers. Let us call the probability distribution $\mathscr{P}$ and whenever we are dealing with a sequence of numbers we shall gather them using face brackets. We then have the notation

$$\mathscr{P}(\mathscr{C}_1, \mathscr{C}_2, \ldots) \equiv \mathscr{P}(\{\mathscr{C}_k\}). \tag{7}$$

The random variables describing the signal, then, are the complex numbers $\mathscr{C}$, and the space over which the signal is defined is the space of the complete set of complex numbers. An element of area in the subspace corresponding to

each complex number can be defined in this way. We define $d^2\mathscr{C}_k$ as the product,

$$d^2\mathscr{C}_k = d(\mathrm{Re}\,\mathscr{C}_k)\, d(\mathrm{Im}\,\mathscr{C}_k). \tag{8}$$

The probability distribution has the following normalization

$$\int \mathscr{P}(\{\mathscr{C}_k\})\, \Pi_k d^2\mathscr{C}_k = 1 \tag{9}$$

when we integrate over the entire space of the complex Fourier coefficients.

So we are beginning to develop a classical noise theory. What happens when we measure the intensity of the field? The intensity of the field is given by the squared modulus of the positive frequency part (say) of the field. That is the same as multiplying the negative frequency part by the positive frequency part,

$$\{|E^{(+)}(\mathbf{r}, t)|^2\}_{\mathrm{av}} = \{E^{(-)}(\mathbf{r}, t)E^{(+)}(\mathbf{r}, t)\}_{\mathrm{av}}. \tag{10}$$

This quantity, however, is a random one, and, of course, if we make any measurement of a random field, we get a random result. To compare with theory, we have to take an ensemble average. This is the meaning of the average we have written. We take the average over the random coefficients $\mathscr{C}_k$ and obtain an intensity. When we measure the intensity of a field we are measuring the ensemble average.

More generally, we can use not just one detector to sample the field, but use two different detectors and superpose their output. In that case we measure an interference pattern which contains the interference of the amplitudes which are brought in by those two detectors, classically. The amplitudes superposed in finding the total intensity can correspond to fields at different spatial positions or delay times. It is thus useful to define the function

$$G^{(1)}_{\mathrm{class}}(\mathbf{r}t, \mathbf{r}'t') = \{E^{(-)}(\mathbf{r}t)\, E^{(+)}(\mathbf{r}'t')\}_{\mathrm{av}}. \tag{11}$$

This is a classical correlation function for the field values at two different space–time points. It becomes the field intensity if $\mathbf{r}$ and $\mathbf{r}'$ are the same and the times t and t' are the same. The field $E^{(+)}$ is linear in the random amplitude parameters $\{\mathscr{C}_k\}$ and $E^{(-)}$ is linear in $\{\mathscr{C}_k{}^*\}$. The ensemble average may also be expressed by

$$G^{(1)}_{\mathrm{class}}(\mathbf{r}t, \mathbf{r}'t') = \int \mathscr{P}(\{\mathscr{C}_k\})\, E^{(-)}(\mathbf{r}t)\, E^{(+)}(\mathbf{r}'t')\Pi_k\, d^2\mathscr{C}_k, \tag{12}$$

where the integration is over the entire space of random coefficients. The study of these random functions of time ($E^{(+)}$, $E^{(-)}$) in which the Fourier coefficients have specified probability distributions is really the subject of stochastic processes, or noise theory. Any such random continuous function of the time is called a stochastic process (a continuous stochastic process in this case). There is a vast body of theory about these processes and there is really no need to go

into further details of that theory here. We have simply noted a few formulae for eventual comparison with the quantum theory.

3. Quantum Theory

In the quantum theory we deal, not with numbers, but with operators. The field is represented by an operator, but that operator can be split into a positive and negative frequency part just as the classical c-number fields. (c-number is a term invented by Dirac for an ordinary number to distinguish ordinary numbers from operators which he called q-numbers.)

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}^{(+)}(\mathbf{r}, t) + \mathbf{E}^{(-)}(\mathbf{r}, t). \tag{13}$$

We are no longer dealing with c-number fields, we are dealing with Dirac's q-numbers, his operators. The negative frequency part of the field is the Hermitian conjugate of the positive frequency part. This is not simply the complex conjugate, but the transposed complex conjugate, i.e. the operator analogue of complex conjugation. Thus, we have

$$\mathbf{E}^{(-)} = (\mathbf{E}^{(+)})^{\dagger} \tag{14}$$

i.e. $\mathbf{E}^{(\pm)}$ are mutually adjoint.

We can make the same mode decomposition of the positive frequency part of the field as we did classically. The operator is expanded in terms of the set of mode functions as discussed above, and the role of the classical Fourier coefficients $\{\mathscr{C}_k\}$ is taken by a sequence of quantum mechanical amplitude operators. By convention, we write these amplitude operators as $\{a_k\}$ and normalize them so that we have $\mathbf{E}^{(+)}$ in the form given by

$$\mathbf{E}^{(+)}(\mathbf{r}, t) = i \sum_k (\tfrac{1}{2}\hbar\omega_k)^{\frac{1}{2}} a_k \mathbf{u}_k(\mathbf{r}) \exp(-i\omega_k t), \tag{15}$$

where $\mathbf{u}_k(\mathbf{r})$ is a vector mode function. The operators a_k are the annihilation operators, one for each mode of the electromagnetic field. For plane waves, a mode of the electromagnetic field is specified by a propagation vector and a polarization for the field which is embedded in the mode function. For example, for plane wave modes

$$\mathbf{u}_k(\mathbf{r}) = \hat{\mathbf{e}}(\mathbf{k}) \exp(i\mathbf{k} \cdot \mathbf{r})$$

where $\hat{\mathbf{e}}(\mathbf{k})$ is the polarization vector. The Hermitian conjugates of the annihilation operators a_k are the creation operators $a_k^{\dagger}$. The normalization factors are inserted to secure the familiar canonical commutation relations between the creation and annihilation operators for the different modes.

$$\begin{aligned} [a_k, a_{k'}^{\dagger}] &= \delta_{kk'} \\ [a_k, a_{k'}] &= [a_k^{\dagger}, a_{k'}^{\dagger}] = 0. \end{aligned} \tag{16}$$

In other words, the one change which we make in going from the classical to the quantum theory is that the complex coefficients (ordinary numbers) become operators and that these operators obey the familiar commutation relations i.e. have the same algebraic properties as the complex amplitude operators which are used to describe the quantum mechanical form of the simple harmonic oscillator. The commutation relations (16) thus define the amplitude operators for an infinite set of oscillators, one for each of the field modes. For harmonic oscillators, it is well known that the products $a_k^\dagger a_k$ have as eigenvalues, the integers $n_k = 0, 1, 2, \ldots$. The integer n_k is just the number of photons in the kth mode.

It is possible to go through a great deal of theory just constructing equations for operators, but, sooner or later, we have to say something about quantum states. Physical quantities are always scalar products of quantum state vectors. We are concerned with states of the radiation field. The ground state of the field is defined as the state $|\text{vac}\rangle$. Here we have used the notation of writing vacuum explicitly in the vacuum state because that is the state in which there are no quanta in any mode. The vacuum state can also be written as

$$|\text{vac}\rangle = |\{0\}\rangle. \tag{17}$$

Here we have the set of quantum numbers zero in all modes of the field. That is to say, if we apply an annihilation operator to the vacuum state, we get absolutely nothing because there are no lower energies available; so applying the annihilation operator annihilates the state thus,

$$a_k |\text{vac}\rangle = 0. \tag{18}$$

We can generate the n quantum states, $|\{n_k\}\rangle$, by applying the creation operators $a_k^\dagger$ to the vacuum state, thus

$$|\{n_k\}\rangle = \prod_k \frac{(a_k^\dagger)^{n_k}}{\sqrt{(n_k!)}} |\text{vac}\rangle, \tag{19}$$

where we see that we construct such a state by creating n quanta in the kth mode, then normalizing, and then doing this for all modes.

We now consider the description of general states of the electromagnetic field. The $|\{n_k\}\rangle$ form a complete orthonormal set (we shall see below some sets which are not normalized and are certainly not orthogonal) spanning the familiar Fock space of quantum field theory. This is a good set of basis vectors with which to describe a general state of the electromagnetic field and, indeed, it was the only one which was ever used in quantum electrodynamical calculations until just a few years ago. Their use was natural enough; in dealing with perturbation theory and never with more than two photons in the field, we might as well confine ourselves to discussion of these particular states,

which are very simple in their orthonormality properties, and very simple in algebraic terms. But the most general state of the field, obviously, is the most general superposition of such n quantum states, and so we have

$$|\ \rangle = \sum_{\{n_k\}} f_{\{n_k\}} |\{n_k\}\rangle. \tag{20}$$

The situation is thus rather different in the quantum mechanical specification of states from the situation in classical theory. The most precise specification in classical theory was achieved by specifying one complex number for every mode of the field. Here this notation indicates a set of coefficients with as many indices as there are modes. If we had one mode only of the field, we should still have an infinite set of coefficients in that specification of a state. For one mode, the state may be expanded as a sum of n quantum states with arbitrary coefficients summed from $n = 0$ to ∞, thus

$$|\ \rangle = \sum_{n=0}^{\infty} f_n |n\rangle, \tag{21}$$

where the only restriction on the f_n is the normalization condition,

$$\sum_n |f_n|^2 = 1. \tag{22}$$

Thus, we see that, in quantum theory, there is an infinite set of complex numbers which specifies the state of a single mode. This is in contrast to classical theory where each mode may be described by a single complex number. This shows that there is vastly more freedom in the quantum theory to invent states of the world than there is in the classical theory. We cannot think of quantum theory and classical theory in one-to-one terms at all. In quantum theory there exist whole spaces which have no classical analogues, whatever.

The correspondence between classical and quantum theory can only be drawn in an asymptotic sense, and usually only in the familiar sense of the correspondence principle of Bohr, which is to say that as we go to very large quantum numbers (i.e. high intensities), we obtain classical results. Now that is a slightly strange statement, familiar as it is, in view of some of the things we have said earlier, because the close correspondence that was observed long ago between classical theory and quantum electrodynamics in the area of optics is really concerned with one-quantum phenomena. In traditional optics, the light beams have been very weak indeed. Zero point vibrations are very strong compared with the strengths of ordinary light beams. (Zero point vibrations may be observed in such devices as the parametric amplifier.) So we have been dealing with a situation in which the fields are actually quite weak. The quantum numbers are very small, and yet that is a situation in which classical theory is applicable. This is a remarkable situation from a quantum mechanical stand point. We should not expect classical theory to

give universally correct answers for situations in which we are dealing with such weak fields. The only way in which quantum theory corresponds to classical theory is, ordinarily, in the limit of large quantum numbers and very strong fields.

4. Intensity and Coincidence Measurements

Let us consider an ideal device, an ideal photon counter. An ideal photon counter is meant to be approximately the simplest thing we can imagine. We also simplify the calculations by showing their skeletal characteristics, by leaving out many of the detailed steps and just trying to convey something of the spirit of the calculation. A photo-detector is a device in which there is, typically, a transition when a photon is absorbed. That may seem trivial but it is a very deep and a very far reaching characterisation of virtually all the photon detection devices that we use. All the practical devices we have, work by absorbing photons from the field and that has some very important consequences for what they detect. In particular, since it is not possible to absorb energy from the vacuum fluctuations, it means, for example, that all of these devices remain completely insensitive to vacuum fluctuations, and, as we mentioned above, that is rather fortunate. As an ideal detector, we shall use a single atom. That is a very good detector, though not a very efficient one, and we might have to work rather hard with the single atom, but the good thing about it is that it is very small compared with the wavelength of visible radiation, and so the chances are very good that we can use the electric dipole approximation to describe a transition. In this approximation, the interaction between the atom and the field is given by

$$H_I = -e \sum_{\gamma} \mathbf{q}_{\gamma} . \mathbf{E}(\mathbf{r}, t) \tag{23}$$

where $\mathbf{q}_{\gamma}$ is the spatial co-ordinate of the γ-th electron relative to the nucleus located at $\mathbf{r}$. Because we are using the electric dipole approximation, it is sufficient to write the electric field operator just at the nuclear position rather than at the position of all the individual electrons. When the atom absorbs a photon, it makes a transition from a certain initial state to a certain final state.

The transition amplitude is given in first order perturbation theory as

$$A_{fi} = M . \langle f | E^{(+)}(\mathbf{r}, t) | i \rangle. \tag{24}$$

There are several little details we have left out, for example, we have paid no particular attention to the conservation of energy in writing down this transition amplitude and we have lumped all of the factors which enter the transition amplitude into the factor M which is the atomic matrix element for going between the intial and final states, whatever they may be. The initial and final state vectors of the electromagnetic field are $|i\rangle$ and $|f\rangle$, respectively. The

operator $E^{(+)}$ is just the positive frequency part of the electric field i.e. $E = E^{(+)} + E^{(-)}$. Here we have just retained the positive frequency part; that is already an approximation we are making. There are two terms in the transition amplitude one of which can easily be resonant and the other one of which never is. The atom, let us say, begins in its ground state, that is in the situation where only absorption takes place. In our detector we begin with very low temperature detectors (all atoms in their ground state). Then all those atoms can do is absorb and therefore only the annihilation operator enters the calculation. That is true because of other properties of the correct expression which we shall come to presently. The conservation of energy is not part of this expression because we have not written down its time integral structure at all. The conservation comes about because we are dealing with functions that oscillate rapidly as a function of time, and as soon as we integrate over a considerable number of cycles of oscillation of the internal motions of the atom and the field, we find we get zero in the integrals unless there is a precise phase matching between the amplitudes of the field and the atomic amplitudes. We shall put in the conservation of energy presently but, for the moment, we do not consider it. It is that which leads us to select only the absorption part (only the annihilation part of the electric field operator) in doing this calculation. If we were dealing with a detection interaction which only lasted one or two atomic cycles, (that would be a ridiculously short time for a detector, 10^{-15} sec or so) then, indeed, we would have to take account of both the positive and negative frequency parts of the field. Usually, we deal with detection processes which are a factor of 10^5 or 10^7 times slower, and so the statement that energy is conserved in the transitions involved has at least an accuracy of one part in 10^5 or 10^7, and it is correspondingly that accurate to discard the negative frequency part of the field.

The transition probability (if you are willing to believe that the transition amplitude is of the form (24), and in its mathematical skeleton it certainly is of that form) is given by

$$T_{fi} = \left\{ |M|^2 \sum_f | \langle f | E^{(+)}(\mathbf{r}, t) | i \rangle |^2 \right\}_{\text{av}} \tag{25}$$

where the sum is taken over all final states because when we detect the photon we make no measurement of the final state of the field. All possible final states of the field can be summed over, but not all conceivable final states of the field are reached in this transition; no matter how many photons there are in the initial state, only one is taken away by the absorption transition. Since we are not observing the final state, we may as well sum over all of the possible final states. All we have said is that most of them do not contribute to the sum but we may as well throw them all in to carry out the sum (mathematically that is

simplest by far). If we do that, we obtain

$$T_{fi} = \left\{ |M|^2 \sum_f \langle i | E^{(-)}(\mathbf{r}, t) | f \rangle \langle f | E^{(+)}(\mathbf{r}, t) | i \rangle \right\}_{\text{av}} \tag{26}$$†

Again the summation is taken over final states. However, the completeness relation for the set of all possible final states is that the sum over final states is just the unit operator, thus we have

$$\sum |f\rangle\langle f| = 1. \tag{27}$$

From (26) and (27) we get

$$T_{fi} = \{ |M|^2 \langle i | E^{(-)}(\mathbf{r}, t) E^{(+)}(\mathbf{r}, t) | i \rangle \}_{\text{av}}. \tag{28}$$

Another way of writing this expression is to think of the outer product (the dyadic product) of the two state vectors as a kind of operator in Hilbert space. We then obtain

$$T_{fi} = |M|^2 \operatorname{Tr} \left\{ \{ |i\rangle\langle i| \}_{\text{av}} E^{(-)}(\mathbf{r}, t) E^{(+)}(\mathbf{r}, t) \right\}. \tag{29}$$

Here we have written the two state vectors $|i\rangle$ and $\langle i|$ side by side and we are now dealing with something which has a square array of components if we take matrix elements. The structure of (29) (precisely the expression (28)) is such that we are evaluating its trace.

We have assumed here that we know the initial state of the electromagnetic field. However, in practice, we rarely have this information. The devices that radiate are always systems with many particles and the most we ever know about them is their statistical behaviour. Thus we are always uncertain of the initial state of the field and the transition probability which we measure is something that is defined only in a statistical sense. We are making a measurement with one atom and we get a particular number for the length of time it takes that atom to make a particular transition in a field; that number will vary from one repetition of the experiment to another. We have to keep repeating the experiment; in other words, we evaluate an ensemble average. That means that we must average (29) over the uncertain initial states of the field. The advantage of the somewhat cumbersome notation of (29) is that we have isolated the two quantities over which we are averaging. We are averaging the dyadic product.

Let us define the density operator, ρ, by the dyadic product averaged over many preparations of the field. We take whatever system it is which produces

† What we have done, is simply to write out the absolute value of the matrix element squared using the familiar theorem that the complex conjugate of a matrix element is the matrix element with the states written in the reverse order of that in which the hermitian conjugate of the operators were taken.

the field and let it produce that field time and time again, thus producing a certain ensemble of initial states of the field. We then average the dyadic product over those indefinite initial states of the field and thus we determine the density operator

$$\rho = \{|i\rangle\langle i|\}_{\text{av}}. \tag{30}$$

The field intensity that we measure is $I(\mathbf{r}, t)$ and is given by

$$I(\mathbf{r}, t) = \text{Tr}\{\rho E^{(-)}(\mathbf{r}, t) E^{(+)}(\mathbf{r}, t)\}. \tag{31}$$

The intensity is the trace of the density operator times the product of the two field operators, the negative frequency part times the positive frequency part. This explicit structure is very important. Notice that the negative frequency part contains all the creation operators, $a_k^\dagger$, when we are quantizing the field and that the positive frequency part contains all the annihilation operators a_k. It follows then that in the vacuum state we get

$$\rho = |\,\text{vac}\rangle\langle\text{vac}\,|$$

$$\text{Tr}(\rho E^{(-)} E^{(+)}) = \langle\text{vac}\,|\,E^{(-)} E^{(+)}\,|\,\text{vac}\rangle = 0. \tag{32}$$

Thus, in the vacuum state the intensity is zero; this is a consequence of the normal ordering of the product of field operators (i.e. the fact that annihilation operators always stand to the right of creation operators). The vacuum state is virtually the only case in which we ever really have a pure state.

The fact that the intensity in vacuum vanishes is very important in avoiding the kind of confusion which arises when the zero-point energy (the zero-point oscillation) is mixed in with observations. If we write expressions other than the normally ordered one, we inevitably run into problems with the zero-point oscillations.

Let us now examine the structure of the kind of statistical average which measures the intensity of the field. Firstly, we shall abbreviate the coordinates by using x instead of a position and time variable. We can define a first order correlation function of the field by

$$G^{(1)}(x_1, x_2) = \text{Tr}\{\rho E^{(-)}(x_1) E^{(+)}(x_2)\}. \tag{33}$$

Here it is defined for two arbitrary space–time points,

$$x_1(\mathbf{r}_1, t_1), \qquad x_2(\mathbf{r}_2, t_2).$$

Had we done the perturbation calculation more fully, including the effects of energy conservation (i.e., putting the time integrals and everything else in) we should have found for the transition probability (the probability that by time t, a single atom undergoes a photoabsorption process) an expression which

looks like this more generally

$$p^{(1)}(t) = \int_0^t \mathrm{d}t' \int_0^t \mathrm{d}t'' \mathscr{S}(t'-t'')\, G^{(1)}(\mathbf{r}t', \mathbf{r}t''). \tag{34}$$

This is still an electric dipole approximation, where $\mathscr{S}$ is a sensitivity factor which contains the atomic matrix elements. We should find that the process is not local in the time variable. There are two different times in this expression (t' and t''). The individual transition amplitude is a time integral and when we square the transition amplitude, the probability obtained is a double time integral. Typically, our device has differing sensitivities for various frequencies of light quanta. All the different sensitivities of the device are easily accommodated in this function $\mathscr{S}$, which can take a variety of forms, and which we shall not discuss here in any detail. (A more complete discussion of photon counting probabilities and sensitivity problems is given in the Les Houches notes, 1964.) For the simplest case of a detector which does not discriminate between frequencies, $\mathscr{S}$ is a delta function.

For this broad band counter, we have

$$\mathscr{S}(t'-t'') \approx s\delta(t'-t''). \tag{35}$$

This cannot be a precise statement because the function s has a one-sided frequency spectrum, but it can be an excellent approximation relative to the relaxation times of the actual field, (i.e. relative to the characteristic times of the function $G^{(1)}$). For this case, with a broad band detector† the integral is a little simpler. The sensitivity is described by a constant and the double integral becomes a single integral.

Then we can think of the absorption process as being local in time, that is, it involves only one time variable.

$$p^{(1)}(t) = s \int_0^t \mathrm{d}t' G^{(1)}(\mathbf{r}t', \mathbf{r}t'). \tag{36}$$

The rate at which the photoabsorption takes place is given by

$$\begin{aligned} w^{(1)}(t) = (\mathrm{d}/\mathrm{d}t)\, p^{(1)}(t) &= s\, G^{(1)}(\mathbf{r}t, \mathbf{r}t) \\ &= s I(\mathbf{r}t) \end{aligned} \tag{37}$$

The electric dipole approximation has neglected the size of the atom and for this reason we have evaluated the field at the position of the nucleus. If we took the size of the atom into account, then we really could not use the electric dipole approximation, and we should probably then have written these cor-

† A broad band detector is one whose sensitivity as a function of frequency is flatter than the frequency distribution of the radiation to be detected. Typically this radiation is in rather a narrow band.

relation functions in terms of the vector potential rather than the electric field operator, but their structure would be identical. We should find, much more generally, that these transition probabilities involve volume integrals over the correlation functions. If we were taking the size of the atom into account then we would have the function $G^{(1)}(\mathbf{r}, \mathbf{r}')$ and we would have to integrate over the variables $\mathbf{r}$ and $\mathbf{r}'$ as well. There would be two more integrals added to the expression and, of course, then there would be the atomic wave functions in the expression. All of these are complications which we really do not need in order to understand the essential points of the theory.

Suppose we have n atoms forming n different broad band detectors with a shutter in front of each atom. We shall open all the shutters at time 0 and let each of them remain open until time t_j where $j = 1, 2, \ldots, n$. The probability, that all n atoms have undergone the photoabsorption transition and the transition has been detected for each of them is given by,

$$p^{(n)}(t_1 \ldots t_n) = s^n \int_0^{t_1} dt_1' \int_0^{t_2} \ldots \int_0^{t_n} dt_n' \times \mathrm{Tr}\{\rho E^{(-)}(\mathbf{r}_1 t_1') \ldots E^{(-)}(\mathbf{r}_n t_n') E^{(+)}(\mathbf{r}_n t_n') \ldots E^{(+)}(\mathbf{r}_1 t_1')\}. \quad (38)$$

This is the sort of structure which can be obtained, formally at least, only in n-th order perturbation theory. But, n-th order perturbation theory is really only an n-fold iteration of first order perturbation theory. There is not really very much added. In other words, to absorb n photons, we require n annihilation operators. We square the matrix element, therefore we have n creation operators (the negative frequency components of the field). We still have the structure we had before in terms of the unknown initial state of the field which has to be averaged over, so we include the density operator. Finally, we have n integrals over n time variables. Each photoabsorption process has been made local in each time variable by the assumption that each atom is a broad-band detector. We may define an n-th order correlation function by

$$G^{(n)}(x_1 \ldots x_{2n}) = \mathrm{Tr}\{\rho E^{(-)}(x_1) \ldots E^{(-)}(x_n) E^{(+)}(x_{n+1}) \ldots E^{(+)}(x_{2n})\}. \quad (39)$$

Again we note that it is a product of $2n$ field operators written explicitly in normal order. The annihilation operators (n of them) standing to the right, the creation operators (n of them) standing to the left. Now, having defined these functions, there are a great many identities we could prove about them, but we won't need any of them for our present purposes. However, one obvious thing to point out is this. We can define a kind of joint counting rate when we have more than one photon counter in the field by differentiating (39) (the probability that by the succession of times $t_1, \ldots, t_n$ all of the atoms have undergone photoabsorption processes which have been detected) with respect to all of the times $t_1, \ldots, t_n$. If we do that, we turn the n-fold integral

into its integrand and obtain

$$w^{(n)}(t_1 \ldots t_n) = \partial^n p^{(n)}(t_1 \ldots t_n)/\partial t_1 \ldots \partial t_n$$
$$= s^n G^{(n)}(\mathbf{r}_1 t_1 \ldots \mathbf{r}_n t_n \mathbf{r}_n t_n \ldots \mathbf{r}_1 t_1). \tag{40}$$

The expression is therefore the n-fold correlation function, but notice that the arguments $\mathbf{r}_1 t_1, \ldots, \mathbf{r}_n t_n$ are repeated. This n-th order correlation function is indeed arrived at in perturbation theoretical terms by an n-th order iteration of first order perturbation theory. But, on the other hand, the result is different from simply the product of first-order perturbation theory taken with itself n times. To put it a little differently, the n-th order correlation function, even with its arguments repeated in this way, does not generally factorize. Physically, this non-factorization is caused by interference effects which take place in the absorption of photons by the different counters. We cannot generally tell which photon is absorbed by which counter and all of the interference terms are implicitly summed in the expression we have written involving the trace with the $2n$ factors of the field. The interference terms would all be omitted if we simply took the product of n first-order correlation functions to represent the right-hand side of (40). The first order correlation function with its arguments repeated is a counting rate for a single counter, and what we are saying is that the n-fold joint counting rate does not factorize into the product of the counting rates that we would measure with each of these counters present in the absence of all the others. In other words, there is generally a tendency towards statistical correlation (correlation, of course, including the idea of anti-correlation as well). There is generally a lack of statistical independence in the counting rates of the n counters in the field.

We can imagine using other sorts of detectors, one possibility suggested by Mandel[2] among others, is to use the process of stimulated emission as a basis for detection. We could put into the field a collection of atoms which are not in their ground state but in the excited state. In this case, the sort of argument we have given would still hold but the first thing we would have to do is to emit photons rather than absorb them and so the operators would occur in anti-normal order rather than normal order, thus

$$p^{(n)}(t_1 \ldots t_n) = s^n \int_0^{t_1} dt_1' \ldots \int_0^{t_n} \mathrm{Tr}\{\rho E^{(+)}(\mathbf{r}_1 t_1) \ldots E^{(-)}(\mathbf{r}_1 t_1)\}. \tag{41}$$

The operators occur in anti-normal order for a detector based on stimulated emission. There are certain practical problems in constructing a detector based on stimulated emission. It is perfectly true that the atoms in an excited state will go to the ground state a little faster when we shine light upon them, and that in principle is detectable, but most of what we would detect would be just the noise of spontaneous emission. The effect of shining the light on these

spontaneously decaying atoms would be rather small so such a detector is not very practical. The point of this argument is simply to emphasise that different operator orderings are associated with different sorts of experiments; it is the experiment which determines the ordering which is convenient. In particular, the stimulated emission device would use anti-normal order. Similarly, we could conceive a device based on scattering. The photons, in being scattered from free charged particles, would make them recoil. Detecting the recoiling particles would be another form of photon detection, a very inefficient one because the cross-sections are small, but one that is used at high energies in the Compton effect detectors. This detector measures a sort of combination which is different again, one which is not at all as simple either as normal ordering or anti-normal ordering. At low frequencies we can probably say that what it detects corresponds more closely to the symmetrical ordering of the annihilation and creation operators. The experiment determines the order. The ordering really does mean something, at least in terms of the convenience of describing particular experiments. Of course, we can always change the orders of operators by using commutation relations but we may increase the complication of the expressions considered by doing that. Typically, for the experiments which have a null result in vacuum, we ought to use normal order; that is the convenient one. For most of what we say, normal ordering will play a particularly important role.

We have not exhausted the possibilities of defining correlation functions. There are many expressions we can define. For example, we can define a new set of correlation functions based on anti-normal order, or any kind of funny order we please. We can, furthermore, define more correlation functions which are based on normal order, and there has been a certain amount of discussion of those, and so let us just write the definition. Here is such a definition,

$$G^{(n,m)}(x_1 \dots x_n y_1 \dots y_m) = \mathrm{Tr}\{\rho E^{(-)}(x_1) \dots E^{(-)}(x_n) E^{(+)}(y_1) \dots E^{(+)}(y_m)\}. \tag{42}$$

This function differs in general from the normally ordered correlation functions we have written earlier by including the possibility of having different numbers of annihilation and creation operators. For $n = m$ we have

$$G^{(n,m)} \equiv G^{(n)}. \tag{43}$$

Expressions of this sort may be defined, but what we find under most of the conditions we study, is that these additional expressions for $n \neq m$ (i.e. the extra ones gained by the definition (42)) typically vanish. Let us see why this is so.

Most optical experiments are performed in essentially a steady state sort of light beam. A steady stochastic process is called a stationary stochastic process.

This does not mean that nothing happens as a function of time, what it means is that all of the statistical statements that we make are independent of the origin of time. That is, our statistical knowledge of whatever time dependent behaviour may be going on is invariant under time displacement. That is what stationarity means for us. Now we can be talking about fields which are oscillating very rapidly; there is no question about the presence of the oscillation, but what we know about the oscillations is independent of the time of day. For example, for a stationary field, the correlation function which generally depends on two times, t_1 and t_2, is invariant under time displacement. This means if we add a time τ to both arguments, we get the same function for all times, τ,

$$\begin{aligned} G^{(1)}(t_1 t_2) &= G^{(1)}(t_1+\tau, t_2+\tau) \\ &= G^{(1)}(t_1-t_2). \end{aligned} \tag{44}$$

That means, in practice, that the correlation function depends only on the difference of the two times. Now, of course, all statistical statements about a stationary field have to be time-independent and that means that there must be a corresponding property for the higher order correlation functions as well. So here, for the general function $G^{(n,m)}$, we may write the same statement

$$G^{(n,m)}(t_1+\tau, \ldots, t_{n+m}+\tau) = G^{(n,m)}(t_1 \ldots t_{n+m}). \tag{45}$$

Now what does that statement mean from a microscopic standpoint? What does it mean about the density operator for the field? Considering any expectation value in the Heisenberg picture, the trace of the density operator times some product of operators (that, of course, is the structure of all our correlation functions) we get,

$$\begin{aligned} &\mathrm{Tr}\{\rho A(t_1+\tau)\, B(t_2+\tau) \ldots Q(t_n+\tau) \\ &\quad = \mathrm{Tr}\{\rho \exp(-iH\tau/\hbar)\, A(t_1)\, B(t_2) \ldots Q(t_n) \exp(iH\tau/\hbar)\}. \end{aligned} \tag{46}$$

Here, the operators are displaced in time. In the Heisenberg picture, we use an exponential based on the Hamiltonian operator to displace operators in time; thus we obtain the right-hand side of (46). So displacement by time, τ, means performing a familiar translation displacement under time, a translation on all of the operators, and when we perform this same unitary transformation on all of the operators, of course all the exponentials cancel except the first one, and the last one. Now, the trace has cyclic symmetry, so the last exponential can be moved around freely to the first position, and the right-hand side of (46) becomes

$$\mathrm{Tr}\{\exp(iH\tau/\hbar)\rho \exp(-iH\tau/\hbar)\, A(t_1) \ldots Q(t_n)\}. \tag{47}$$

All of the remaining operators are evaluated at their original times $t_1, \ldots, t_n$.

We can write (47) in the form

$$\mathrm{Tr}\{\rho A(t_1) \ldots Q(t_n)\}, \tag{48}$$

provided the transformed version of the density operator is equal to the density operator itself. In other words, provided the Hamiltonian commutes with the density operator,

$$[H, \rho] = 0.$$

Commutation of the density operator with the Hamiltonian is the necessary and sufficient condition, finally, for stationarity.

Now supposing we have a stationary field, let us see what we can say about the correlation functions, and, in particular, these correlation functions with different numbers of creation and annihilation operators. Suppose the field is one in which the set of modes which we label by "k" is occupied. The first of the more general correlation functions may be constructed as one in which we have no creation operators, and one annihilation operator. It then just has one time argument. This function is, of course, the trace of ρ times $E^{(+)}$ at the time t. It is just the expectation value of the field. The form it takes is necessarily this,

$$G^{(0,1)}(t) = \mathrm{Tr}(\rho E^{(+)}(t)) = \sum_k F_k \exp(-i\omega_k t). \tag{49}$$

If we have certain modes of the field which are occupied, we imagine the mode decomposition of the operator $E^{(+)}$ and calculate the expectation value and, indeed, we have a sum of periodic terms which can only be the exponentials associated with the time dependences of the occupied modes. Now, we are assuming that the field is stationary, and if the field is stationary, we simply cannot have oscillations like that. Everyone of those coefficients F_k must vanish. If the field is to be stationary, the only possibility is that the correlation function $G^{(0,1)}(t)$ vanishes. That is not surprising. For a stationary field, the expectation value of the field amplitude itself must vanish, so the first of these more general correlation functions vanishes.

Now, how about the other correlation functions? It is always possible that we have equal numbers of creation and annihilation operators to pair-off the oscillating time-dependencies and in that way to get a time-independent function. However, if we have unequal numbers of creation and annihilation operators, we are dealing with a condition which is very much more difficult to satisfy. The more general situation for a field with $\{k\}$ modes occupied is,

$$G^{(n,m)}(t_1 \ldots t_{n+m}) = \sum_{k_1} \cdots \sum_{k_{n+m}} F_{k_1 \ldots k_{n+m}} \exp\left[i\left\{\sum_{j=1}^{n} \omega_j t_j - \sum_{l=n+1}^{n+m} \omega_l t_l\right\}\right]. \tag{50}$$

In general, the (n, m)th order correlation function is a summation for which in all the terms of the summand, there occur all possible choices of the expo-

nential time dependencies which characterize the modes: there appears in the positive frequency sense, m such factors, and in the negative frequency sense there appear n such factors. If there is any unbalanced oscillation in these two exponents, and if we are asserting that the field is stationary, viz. that this function is independent of time displacements, then the only way that that can be secured is to say that the corresponding coefficient vanishes. A necessary condition, then, to have a non-vanishing function $G^{(n,m)}$ is that it be possible to put together sets of frequencies of n frequencies and m frequencies, (and we can allow repetitions), so that

$$\sum_{j=1}^{n} \omega_j = \sum_{l=n+1}^{n+m} \omega_l. \tag{51}$$

We take the set of occupied modes, write down their frequencies, and choose n of them with possible repetitions, and choose m of them allowing for possible repetitions. However, if it is not possible to do that so that the one sum is precisely equal to the other, then there is going to be unbalanced oscillation present in the corresponding term for the correlation function, and that coefficient must vanish if the field is to be stationary. That means that the simplest way of securing a non-vanishing correlation function for a stationary field is just to have $m = n$ and to make identical choices of the sets of frequencies. Then the one time dependence will cancel the other and it is very easy to have a non-vanishing function. It is not so easy if we choose $m \neq n$. To satisfy a condition of this sort a very special set of frequencies must be occupied in the field. For example, if we have three frequencies present, then we see that if we call them ω_1, ω_2 and ω_3 we must have some such relationship as this,

$$\omega_1 + \omega_2 = \omega_3, \tag{52}$$

among the three frequencies, if we are to get a non-vanishing result for what we have defined as $G^{(1,2)}$. In fact, a condition of this sort can hold, for example, in the parametric amplifier, or in the coherent Raman effect where the modes excited have this sort of property. But this is only a necessary condition in order to have a non-vanishing correlation function for these peculiar orders. It is only a necessary condition because, in fact, if these modes at different frequency oscillated independently of one another, we should have to look further at the structure of the coefficients, and then we still should not find any which were different from zero under the stationarity condition. There has, additionally, to be a statistical correlation between the density operators for these particular modes. If, on the other hand it were true that the density operator for the entire field factored into a product of density operators (stationary density operators for the individual modes) once again, we could show that the functions vanish. The actual physical situation in devices like the parametric amplifier or a medium in which the coherent Raman effect is

amplified, or a substance in which second harmonic production is occurring, is one in which there do exist such statistical correlations between the modes of different frequencies. And so, it is true that these peculiar correlation functions take on non-vanishing values.

In the case of harmonic production which is the simplest, we send in red photons, and pairs of red photons join together to form blue photons. In the outgoing field we have two components present, one of which is at precisely twice the frequency of the other. Furthermore, there are phase relations between the two fields. There is statistical dependence between these two fields, and, in fact, in the region illuminated by such a crystal producing the second harmonic, we should find that the function $G^{(1,2)}$ is different from zero. But it takes all of the complication of non-linear optics to bring that situation about. If we just produced two fields from independent lasers, say, which happened to have frequencies such that the one had precisely twice the frequency of the other, the function $G^{(1,2)}$ would still vanish, because there would then be no phase relations between the two fields. The dynamical process of harmonic generation is, here, precisely that which does generate the phase relations to give a non-vanishing function.

It is interesting to consider how we would detect the function $G^{(1,2)}$. We would have to perform a curious sort of interference experiment in which an amplitude based on $(E^{(+)})^2$ was somehow or other able to interfere with an amplitude based on a single factor of the field $E^{(-)}$. It is not impossible to do that and we may consider the following experiment as a suggestion as to how it may be done. Figure 1 shows a device which is producing harmonics. The problem is to make the two beams R, B interfere. There is a variety of ways. We can engage in harmonic production once more on the red beam. Of course, we shall then get some of the blue light through and a little bit more. The red light will go through and produce some blue light. This is a species of interference experiment because we can now let the blue beams interfere with one another. However, if we do that, we are measuring $G^{(2)}$ which is something quartic in the field and it is not what is wanted. This $G^{(2)}$ is the average of $(E^{(-)})^2\,(E^{(+)})^2$. What we have to do is some kind of experiment which literally measures the high frequency field against the low frequency field. It has to be an interference experiment of some sort. A possibility is the following. Suppose we use an atom as a photon detector and examine the photoelectrons emitted. Suppose also that atom has a threshold for photon detection which lies higher in frequency than the red frequency used. A single red photon will not give a photo-electron, but two red photons will. Now, two photons *can* be observed simultaneously and here we have a good strong beam of red photons, so there will be some photoelectric effect and that photoelectric effect will occur only in second order; it will be proportional to $(E^{(+)})^2$. Alternatively, the blue photons, which have, each of them, twice the frequency, can make the photo-

electric transition occur by the absorption of a single quantum. These two processes are going to interfere with one another, and the amplitude of the interference term for the two processes will be proportional to the statistical average of $E_B^{(-)} E_R^{(+)} E_R^{(+)}$, i.e.

$$\mathrm{Tr}(\rho E_B^{(-)} E_R^{(+)} E_R^{(+)}).$$

As we move the photodetector which is based on the interference of two photon absorption versus single photon absorption, we shall discover interference terms just because of a small difference of propagation vectors which typically occurs. The propagation vector of the blue photons will not be

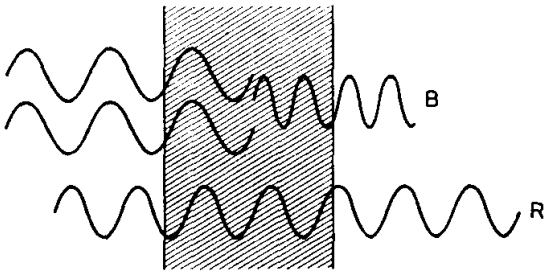


FIG. 1. Second harmonic production in a medium; some red light is converted to blue light; some red light is transmitted.

precisely equal to twice the propagation vector of the red photons ($k_B \neq 2k_R$ in general) because of dispersion. Therefore, as we move the detector based on the interference of these two different sorts of absorption processes, we shall find fringes in the interference of the two absorption processes. So, in principle, this is a possible measurement.

Another possibility is to have two converters as shown in Fig. 2. Many red photons enter the system, some are converted to blue ones, and some remain red. We have a second converter located as shown and some of the remaining red ones are turned blue. Finally, we have a blue filter, which removes the red. An ordinary photodetector is used. The total field between the two converters is the sum of the blue and red components. The final intensity measurement is $G^{(1)}$ in the region beyond the converters, and is given by

$$G^{(1)} = \mathrm{Tr}\{\rho E''^{(-)} E''^{(+)}\}. \tag{53}$$

This expression is quadratic in the field E''. The field E'' in the region beyond the second converter contains the more-or-less unchanged field E_B' and a part which is proportional to the square of the red component, thus we have

$$E''^{(+)} = E_B'^{(+)} + \text{const}\,(E_R'^{(+)})^2. \tag{54}$$

If we substitute the sum of these two terms for the doubly primed fields in the intensity we measure, we see that the function $G^{(1)}$ contains $E_B'^{(-)}$, $E_R'^{(+)}$,

i.e. $G^{(1)}$ contains

$$\mathrm{Tr}\{\rho \mathrm{E_B}'^{(-)} E_{\mathrm{R}}'^{(+)} E'_{\mathrm{R}}{}^{(+)}\}.$$

In other words, $G^{(1)}$ contains the function $G^{(1,2)}$ when we are using fields between the two converters. There are other ways of constructing the intensity measured finally. For example, we can go back to the original fields and write the intensity in terms of those, but then it becomes a quartic expression. So it really all depends on which fields we are considering, whether we are measuring one order of the correlation function, or another. Viewed in these terms, the measurement made is one of the (2, 2) correlation function, or the ordinary 2nd order correlation function when dealing with fields before they have entered

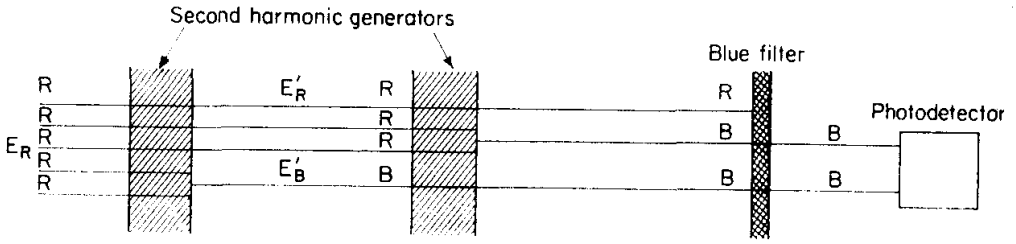


FIG. 2

the system. For fields before entering the converters, $G^{(1)}$ contains the second order correlation function,

$$\mathrm{Tr}\{\rho E^{(-)} E^{(-)} E^{(+)} E^{(+)}\} = G^{(2,2)} = G^{(2)}. \tag{55}$$

So in this sense, it is a matter of definition, just which of these correlation functions we are considering. But if we are dealing with the fields in between the two converters, then we can view the combination of the last converter, the filter, and the photodetector as being equivalent to the single atom photodetector with the high threshold which we considered above.

We have indicated that the higher order correlation functions with the differing numbers of creation and annihilation operators can, indeed, be different from zero and be measurable. However, this is not done to give too heavy an emphasis to these curious sorts of correlation functions; in fact under all ordinary experimental circumstances in stationary fields (whenever we have a steady beam, we effectively have a stationary field) for $m \neq n$, they vanish. Therefore, it is quite sufficient to retain the simpler set of correlation functions and deal just with those i.e. the ones for which the number of annihilation operators is equal to the number of creation operators. In practice those are almost always the functions which we measure.

5. Coherence

We have not said a word about coherence yet, so let us go back to the most elementary considerations and look at the first sort of experiment in which we would propose to use the term coherence. This goes way back to optics. Figure 3 is a crude representation of Young's experiment. The screen has two pinholes which are very tiny because we do not want to calculate the diffraction pattern of each hole. For all we care, the wave which spreads behind each of these holes is perfectly isotropic. We are looking at the intensity on a distant

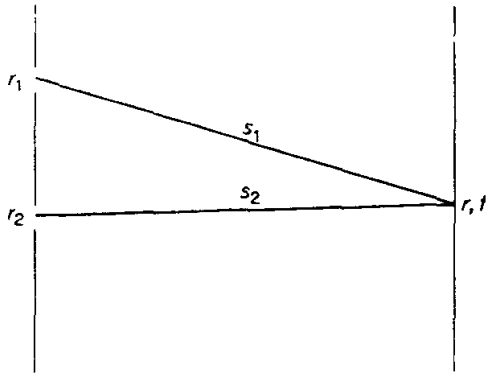


Fig. 3. Young's experiment.

screen at a point r, t. The field at r is, to a good approximation by Huygens' principle, a superposition of the fields at these two pinholes.

The point considered lies at two different distances from the two pinholes and so we are effectively sampling the fields at these two pinholes at two different times. The distances are s_1 and s_2 and the times are $[t-(s_1/c)]$ and $[t-(s_2/c)]$. The field is given by

$$E^{(+)}(r,t) = \lambda\left\{E^{(+)}\left(r_1\, t-\frac{s_1}{c}\right)+E^{(+)}\left(r_2\, t-\frac{s_2}{c}\right)\right\}. \tag{56}$$

All of diffraction theory is bound up in the parameter λ which tells us the transmission of the hole. (We are assuming that this is independent of the angle at which these propagation vectors lie on the other side of the hole). We are taking the positive frequency component of the field. Abbreviating the appropriately retarded space–time points by X_1 and X_2, we get

$$E^{(+)}(r,t) \equiv \lambda\{E^{(+)}(X_1)+E^{(+)}(X_2)\}. \tag{57}$$

The intensity is found from the correlation function by evaluating the fields $E^{(+)}$ and $E^{(-)}$ at the same space–time point which is the space–time point on the screen, thus

$$I(r,t) = \mathrm{Tr}\{\rho E^{(-)}(rt)\, E^{(+)}(rt)\}. \tag{58}$$

But each of these fields is a superposition of the field operators (i.e. a sum)

corresponding to the appropriately retarded times at the pinholes. Substituting the two sums, we get

$$I(r,t) = |\lambda|^2 \operatorname{Tr}\{\rho(E^{(-)}(X_1)+E^{(-)}(X_2))(E^{(+)}(X_1)+E^{(+)}(X_2))\}. \quad (59)$$

This is a sum of correlation functions. Apart from the constant λ^2 which contains all the physical optics, it is the sum of four correlation functions,

$$I(r,t) = |\lambda|^2\{G^{(1)}(X_1,X_1)+G^{(1)}(X_2,X_2)+G^{(1)}(X_1,X_2)+G^{(1)}(X_2,X_1)\}. \quad (60)$$

The function $G^{(1)}(X_1, X_1)$ is the intensity obtained if the second hole is closed. The function $G^{(1)}(X_2, X_2)$ is the intensity if the first hole is closed. Then we have two terms which exist only because both holes are open. (We cannot tell which hole any given photon went through). These two terms are complex conjugates of one another so they are just twice the real part of $G^{(1)}$ with the arguments in either order,

$$G^{(1)}(X_1, X_2)+G^{(1)}(X_2, X_1) = 2 \operatorname{Re} G^{(1)}(X_1, X_2). \quad (61)$$

If we write the function as its absolute value times an exponential of unit modulus (i.e. $e^{i\phi}$), then the real part is just $|G^{(1)}(X_1, X_2)|$ times the cosine of the phase factor ϕ,

$$I(r,t) = |\lambda|^2\{G^{(1)}(X_1,X_1)+G^{(1)}(X_2,X_2)+2|G^{(1)}(X_1,X_2)|\cos(X_1,X_2)\}, \quad (62)$$

where we have taken $G^{(1)} = |G^{(1)}|\, e^{i\phi}$. Let us now consider what happens as we move the point of observation up and down the screen. The positions of the holes at r_1 and r_2 (the spatial parts) of the space–time position X_1 and X_2, remain fixed. As the two distances s_1 and s_2 vary the times at which we sample the fields vary. The absolute value remains positive and represents the envelope of the sort of wiggle which is associated with the cosine. The wiggle represents the fringes; the intensity goes up and down. The rising and falling of the intensity (i.e. the degree of contrast) is governed by $|G^{(1)}(X_1, X_2)|$. The function $G^{(1)}(X_1, X_2)$ is a correlation function. If it vanishes, it means that the fields at the two pinholes are uncorrelated. If there is no correlation, we say that the light at the two points is incoherent at the one point relative to the other (i.e. relatively incoherent). On the other hand, increase in coherence results in an increase in fringe contrast, i.e. the amplitude of the wiggles becomes large. Since $|\cos\phi|$ is never greater than unity, the fringe contrast is maximised by making $|G^{(1)}(X_1, X_2)|$ as large as possible, and that signifies maximum coherence. There is an upper bound because the total intensity of the expression can never become negative. It is very important that we have the intensity terms $G^{(1)}(X_1, X_2)$ and $G^{(1)}(X_2, X_1)$ because they are intrinsically positive whereas the interference term oscillates between positive and negative

values. The upper bound to $|G^{(1)}(X_1, X_2)|$ is given by the Schwarz inequality,

$$G^{(1)}(X_1, X_1)\, G^{(1)}(X_2, X_2) \geqslant |G^{(1)}(X_1, X_2)|^2. \tag{63}$$

This inequality can be proved easily from the positive definite character of the density operator; the fact that the density operator can never have negative eigenvalues. The squared modulus of the cross-correlation function has to be less than, or equal to the product of the two intensities. We secure maximum fringe contrast by using the "equals" sign.

$$G^{(1)}(X_1, X_1)\, G^{(1)}(X_2, X_2) = |G^{(1)}(X_1, X_2)|^2. \tag{64}$$

That is a perfectly good characterization of optical coherence as it has always been used in all of the older contexts of optics. Let us agree that that is what we mean by optical coherence.

The fields are coherent at two space–time points (X_1, X_2) if the equality holds and, furthermore, we have optical coherence of the entire field if this equality holds for all space–time points X_1 and X_2. This is a slightly peculiar looking condition, and it is not immediately obvious how it is satisfied. Let us cite one *sufficient condition* for satisfying the optical coherence condition. We shall call the optical coherence condition (64) the first order coherence condition because it deals only with the first order correlation function. We have already seen that there is a hierarchy of correlation functions and we shall talk about other sorts of optical coherence in connection with those below. Suppose that the first order correlation function factorizes in the form,

$$G^{(1)}(X_1 X_2) = \mathscr{E}^*(X_1)\, \mathscr{E}(X_2). \tag{65}$$

If we write it in any factored form and then use the fact that the function with its arguments reversed is the complex conjugate of the function, then we see immediately that the two factors must be complex conjugates of one another. Furthermore, the field $\mathscr{E}$ which represents one of the factors must be a positive frequency field, that is $\mathscr{E}$ contains $\exp(-i\omega_k t)$. If this factorization condition holds, then by just noting that $G(X_1, X_1)$ is the squared modulus of this field and $G(X_2, X_2)$ is likewise, we see that the condition for maximum fringe contrast holds identically. Obviously, this is a sufficient condition for optical, or first order, coherence. What is a good deal less obvious, but nonetheless true, is that if the maximum fringe contrast condition holds everywhere in space–time, then *it* implies the factorization condition. It does not take any deep mathematics to prove that, it is just the Schwarz inequality but it does take about ten minutes to prove. It is proved in the Les Houches notes, and it is also proved by Titulaer and Glauber[3]. This definition of optical coherence goes a little bit beyond the definition that had earlier been used in optics because of the usual limitation of experimenting with stationary light beams, in fact, the usual definition of the correlation function in optical terms was

made in terms of a time average rather than an ensemble average. If we are dealing with a time average, we have only one time left in the expression, so it is not easy to see then how to extend the definition to deal with non-stationary fields. The new definition is a definition which extends immediately to non-stationary fields. It is also a quantum mechanical one rather than a classical one.

One of the nice properties of the statement of factorization, once we know that the statement of factorization is equivalent to the statement of maximum fringe contrast, is that we can see clearly what the relationship is between monochromaticity and optical coherence. As we know, all of the efforts over many years to increase optical coherence were efforts to provide more and more monochromatic optical fields. The curious thing about the definition we have given here is that it says absolutely nothing about monochromaticity. In fact these fields $\mathscr{E}$ into which the correlation function factorizes can have absolutely arbitrary spectra. So an optically coherent field according to our definition need not be monochromatic at all. However, when we restrict ourselves to dealing with stationary fields, then indeed, the correlation function must be invariant under time displacements, i.e. it must depend only on the difference of two times

$$G^{(1)}(t_1, t_2) = G^{(1)}(t_1 - t_2) = \mathscr{E}^*(t_1)\,\mathscr{E}(t_2). \tag{66}$$

If we require then that it factorizes as well, that is, if we impose the first-order coherence condition, then the only solution of this functional equation (the only way of writing a product which depends only on the difference in the two times) is to say that the field $\mathscr{E}$ depends exponentially on the time, and then since it must be a positive frequency field, it can only take the form

$$\mathscr{E}(t) = \mathscr{E}(0) \exp(-i\omega t). \tag{67}$$

Are there any fields that obey our optical coherence condition precisely? One trivial sort of example is just a classical field which is perfectly well determined in its behaviour, one whose Fourier coefficients are absolutely fixed. Then the procedure of averaging over unknown Fourier coefficients referred to above becomes a trivial one and it is clear that the correlation function factorizes in this case. Of course, we are dealing there with the classical correlation function and it is that that factorizes. We have said nothing about the quantum mechanical one and we leave open for the moment the question of whether there are quantum mechanical fields which do this; as we know, there is a vast variety of them. The reason we mention this particular classical example is to show the first of the many associations we shall see between the definiteness of the field, the fixed character of the (otherwise) random Fourier coefficients and coherence. Coherence tends to limit the noise in the field. The more coherence, we shall see, the less noise is allowable. Optical coherence is a rather weak limita-

tion on noise in the field. We can have quite noisy fields which are still consistent with the optical coherence condition and only by adding further conditions can we get rid of much more noise.

We can go on; having defined first order coherence, and being equipped with a great many correlation functions, we can define more exotic sorts of coherence. An obvious thing to do is to look at the higher order correlation functions and require that they factorize in essentially the same way. Let us assume that the first m correlation functions 1, ..., m all factorize in essentially this way,

$$G^{(m)}(X_1, \ldots, X_{2m}) = \mathscr{E}^*(X_1) \ldots \mathscr{E}^*(X_m)\, \mathscr{E}(X_{m+1}) \ldots \mathscr{E}(X_{2m}), \tag{68}$$

for $m \leqslant n$, where the function $\mathscr{E}$ is the same in all cases and is not an operator, it is an ordinary function which is a positive frequency solution of the Maxwell equations that obeys all the boundary conditions which the field must obey. A field which obeys this condition for the first n correlation functions will be called nth order coherent. There is room for some variation in the choice of the particular definition of nth order coherence; some other suggestions have been made. Cases in which we have, in practice, fields with precisely 17th order coherence (say), and no higher coherence are not known; it is intended to suggest by this definition that we are adding more and more conditions and, indeed, when we let $n \to \infty$, we shall then be talking about a fully coherent field; that is the other significant extreme. If nth order coherence holds, then (68) implies that all of the correlation functions which represent joint counting rates for m-fold coincidence experiments and $m \leqslant n$ factorize in this simple way:

$$G^{(m)}(X_1, \ldots, X_m, X_m, \ldots, X_1) = \prod_{j=1}^{m} G^{(1)}(X_j, X_j). \tag{69}$$

Notice that the arguments are repeated here, X_1 occurs twice, and finally X_m occurs twice. In other words, fields with higher order coherence are the fields for which the registry of counts by the individual photon counters *is* statistically independent. We put n different counters in the field and they will each record photons in a way which is statistically independent of all of the others with no special tendency towards coincidences, or correlations, or anti-correlations, except as is governed by the overall intensities of the fields at those particular points. (Of course, if the field strength essentially goes to zero all over the place, none of the counters will register.) Here we are considering correlation, not rates, and there will be no tendency towards correlation, given higher order coherence.

Now let us switch away for a moment and say why it is interesting to define such a curious idea. In the Hanbury-Brown–Twiss experiment[4] (Fig. 4) light from a discharge tube (a narrow bandwidth spectral source) is passed

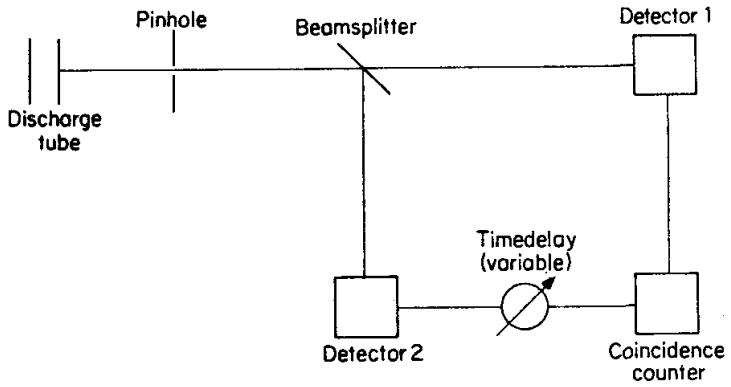


FIG. 4. Hanbury-Brown–Twiss experiment.

through a pinhole and emerges as a very coherent beam in the first order sense. The beam is divided by a half-silvered mirror and recorded by two photodetectors. In the original experiment these were photomultiplier tubes which gave a more-or-less continuous output which was random. The two outputs were then multiplied together to see if there was any correlation between them and such a correlation was found. In all later versions of this experiment, the detectors are photon counters and coincidence detection is used. In the original experiment the detectors were moved relative to their zero points and in all subsequent versions of the experiment, the counters are kept more or less fixed and variable time delay is used in the coincidence gating. In this experiment the coincidence rate was plotted against time delay (the two counters have finite resolving times and so we expect some smearing-out of coincidences). We should expect that if the entry of the photons into the two counters were statistically independent events we should see a uniform background in the coincidence rate versus the time delay (Fig. 5). In the original experiment a very tiny deviation from the uniform coincidence rate versus time delay was observed. In fact, however, the effect is not a tiny one. If it is possible to get the band width of the incident radiation to be narrow enough,

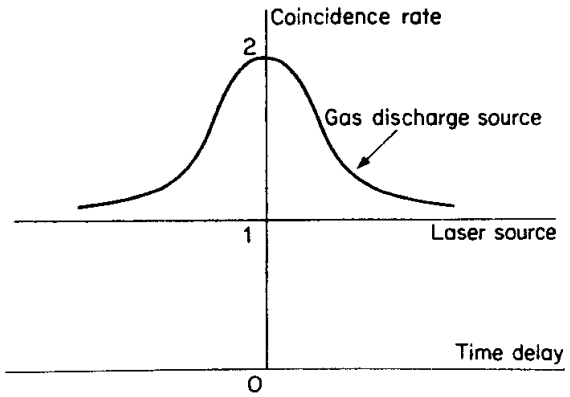


FIG. 5. Results of Hanbury-Brown–Twiss experiment.

in other words, if the experiment is done in ideal circumstances, we find a coincidence rate corresponding to zero time delay which is twice the accidental background rate. So there is in the light entering the system, a distinct tendency for photons to occur in pairs, to enter the system in pairs, or at least for these two photon counters to register photons simultaneously. That is the Hanbury-Brown–Twiss effect (HBT effect).

Why do we see this effect? Is it, for example, some trick in the mirror? No, the only function of the mirror is to allow us to do something which would otherwise be very inconvenient, which is to put two counters at the same place in space. The mirror creates the same field in front of counter 1 as is in front of counter 2. We could perform the experiment with a single counter if we had a fast enough resolving time. We should investigate the distribution of intervals between photon counts and we should discover that there is an anomalously large number of short intervals between photon counts. There is again a distinct tendency for photons to clump as they enter a given counter. Well, here we have two photon counters in the field and they show results which are not statistically independent. This is a situation in which $G^{(2)}$ is definitely not equal to $G^{(1)}G^{(1)}$. There is no second order coherence. Then we might think that there is no sense in defining such a notion. But, in fact, this statement is not true for all fields and one of the ideas which led to all the work we have been describing is the surmise, very quickly born out by experiment, that in fact there is no HBT effect when a stabilized laser beam is used as the source. When we plot the coincidence rate, we find no bump at all (Fig. 5), regardless of the time delay. No HBT effect occurs in a stablized laser beam. Well, that is the sense of defining it. There is now a good deal of verification that the factorization holds, certainly up to some quite high order (six) of the correlation functions. So up to sixth order there is this factorization in the stabilized output of a high quality gas laser; that is the rationale in defining these correlation functions and discussing higher order coherence.

Let us now consider a little further the consequences of defining higher order coherence. We can prove some mathematical theorems and if we return to the papers of Titulaer and Glauber[3] one of the interesting things is this. There are more consequences of the first-order coherence notion (i.e. maximum fringe contrast, or the factorization condition, which are equivalent) than first appears. One consequence is that a species of factorization holds in all of the higher order correlation functions. They all factorize into approximately the form we should like but with a coefficient g_n which is generally different from unity.

$$G^{(n)}(X_1, \ldots, X_{2n}) = g_n \mathscr{E}^*(X_1) \ldots \mathscr{E}^*(X_n)\, \mathscr{E}(X_{n+1}) \ldots \mathscr{E}(X_{2n}). \tag{70}$$

In higher order coherence the coefficients g_n (which can generally be any

positive numbers) are all equal to 1,

$$g_n = 1, \quad \text{all} \quad n.$$

So we can specify the coherence properties of a beam by considering first the factorization of the first order correlation function and then, if it factorizes, by discussing the discrete sequence of numbers g_n (they are quite measurable numbers). A problem in all definitions of coherence is that they can only be fulfilled in approximate terms. If we speak of factorization as a coherence condition, in actual fact, we should never fulfil those conditions everywhere in the universe. But a laser beam can do it over vast intervals of the space variable and intervals of the time variable, perhaps as long as 0·1 sec.

There are a number of other consequences of the assumption of first order coherence. Let us consider the coherence conditions in still one more way. One of the ways of securing the first order coherent beam is to occupy just one mode. If only one mode of the field is occupied then we can always write the correlation function in factorized form, so one mode occupation does it. That will always allow us to write $G^{(1)}$ as a factorized form. We can broaden that example greatly if we define modes more generally than we are accustomed to, e.g. define a set of mode functions $v_l(r, t)$ in terms of our other mode functions $u_k(r)$, as

$$v_l(r, t) = \sum_k \gamma_{lk} u_k(r) \exp(-i\omega_k t). \tag{71}$$

The new set of mode functions are non-monochromatic mode functions. It is easy to define, as linear combinations of the familiar creation and annihilation operations, new creation and annihilation operators for these new non-monochromatic modes. These non-monochromatic modes are perhaps also describable as wave packets (whether we should call them modes is open to question). The most general first order coherent field is one in which only one of these generalized modes, or only one of these wave packets has any photons in it at all, other varieties are empty. That is a fairly simple characterization of first order coherence.

All coherence, or higher and higher order coherence represents more and more constraints upon the distribution of quantum numbers for the occupation of that one mode, and, finally, full coherence, implies that there is a Poisson distribution for the number of photons. These are the definitions of coherence. Again, are there any fields which will fill all of them? There is the trivial example of a classical field with no uncertainty in it. That is a classical field in which the Fourier coefficients are absolutely fixed. For such a field, all of the classical correlation functions factorize in just the way we are describing. So now it is obvious that any noiseless field, any perfect signal, is fully coherent. What our hierarchy of definitions of coherence has done is to approach in the limit, the sort of field which any communication engineer

would say is coherent, viz. one which involves no noise, except, finally, the trivial sort of noise, which represents a phase arbitrariness in the field $\mathscr{E}$. Any overall phase in the expression just cancels out. So much for the subject of coherence.

6. Coherent States

We shall now consider quantum mechanical fields which have the property of full coherence. First we shall consider pure states since these are likely to be the simplest. That means that the density operator can be written as a trivial dyadic product of this state with itself,

$$\rho = |\rangle\langle|. \tag{72}$$

Then our correlation function $G^{(n)}$ takes the form of an expectation value in that state of a normally ordered product of operators with creation operators on the left, annihilation operators on the right,

$$G^{(n)} = \langle| E^{(-)} \dots E^{(+)} |\rangle. \tag{73}$$

What kind of pure state leads to factorization of this expression? If it were possible to diagonalize the operators $E^{(+)}$ and $E^{(-)}$ simultaneously, then such an eigenstate of them would fill the bill very nicely, but we cannot do it because the operators $E^{(+)}$ and $E^{(-)}$ do not commute. Suppose we continue the restriction of dealing with just those experiments which can be described in terms of normally ordered products. As long as the products are normally ordered, we do not really need simultaneous eigenstates of $E^{(+)}$ and $E^{(-)}$ because all we ever find is $E^{(+)}$ at the right and $E^{(-)}$ at the left so it is quite sufficient to find an eigenstate in this sense. The $E^{(+)}(r, t)$ applied to our state gives an ordinary function, $\mathscr{E}(r, t)$ multiplied by that state,

$$E^{(+)}(r, t)\,|\rangle = \mathscr{E}(r, t)\,|\rangle. \tag{74}$$

Of course, if that is true, then there will be a dual relationship which is this: the dual state with the Hermitian adjoint operator applied will give the complex conjugate eigenvalue $\mathscr{E}^*(r, t)$ field multiplied by the dual state,

$$\langle| E^{(-)}(r, t) = \mathscr{E}^*(r, t)\,\langle|. \tag{75}$$

With these two relationships, we have secured, for all normally ordered products, an expectation value which factorizes, and therefore $G^{(m)}$ will be given by

$$G^{(m)} = \mathscr{E}^*(r_1, t_1) \dots \mathscr{E}(r_{2m}, t_{2m})\,\langle|\,|\rangle. \tag{76}$$

The two relations (74) and (75) together provide a sufficient condition for factorization. We have not in this way determined the most general set of pure states which does this. That is done in the papers of Titulaer and Glauber.[3]

The additional states that are found are not very interesting. They differ just in having some extra phase factors, which the states we shall discuss do not contain. The additional ones are ruled out by considering the more general set of correlation functions $G^{(n,m)}$. If we require that these factorize as well, the only pure state that will do it is the state that satisfies the eigenvalue condition. The eigenstates of $E^{(+)}(r,t)$ are the coherent states. It is obvious that the states of (74) are not states with a fixed number of quanta in them because $E^{(+)}$ is an annihilation operator and the statement here is that we are applying an annihilation operator taking away one photon and, even so, we have the same state left. That is not possible if we have a fixed number of photons, so the number of photons must be indefinite. Now to see what the eigenstate condition (74) means in greater detail let us expand the positive frequency field operator in terms of normal modes as we did in (15)

$$E^{(+)} = i \sum (\hbar\omega_k/2)^{\frac{1}{2}} a_k u_k(r) \exp(-i\omega_k t). \tag{77}$$

A corresponding expansion must be available for the eigenvalue function

$$\mathscr{E} = i \sum (\hbar\omega_k/2)^{\frac{1}{2}} \alpha_k u_k(r) \exp(-i\omega_k t). \tag{78}$$

Since the mode functions $\mathbf{u}_k(r)$ form an orthogonal set the eigenstate condition (74) implies that

$$a_k|\,\rangle = \alpha_k|\,\rangle \tag{79}$$

for all values of the mode index k. Since it is possible to project out of the dependence on the spatial coordinate, r, all the various coefficients associated with the different mode functions, then it must obviously be true that a_k applied to this unknown state gives us the number α_k applied to the unknown state. In other words, these states are the right eigenstates of all of the annihilation operators and now we know the properties of those states. Equation (79) alone is sufficient to determine them for us. For one mode, it follows from this equation, that the state can be expanded in the following way in terms of the n quantum states,

$$|\alpha\rangle = \exp(-\tfrac{1}{2}|\alpha|^2) \sum \frac{\alpha^n}{\sqrt{n!}} |n\rangle \tag{80}$$

where the normalization factor is $\exp(-\frac{1}{2}|\alpha|^2)$. Then we label this one mode state with the complex eigenvalue itself.

Now, obviously there is such a state for any one mode for any complex number α that we choose. For the full set of modes we can write

$$|\{\alpha_k\}\rangle = \prod_k |\alpha_k\rangle_k. \tag{81}$$

We secure all possible classical solutions to the Maxwell equations by substituting in (80) all possible complex coefficients α_k and, therefore, we see that

there is a one-to-one correspondence between eigenstates of (74) (i.e. eigenstates of the positive frequency part of the field) and classical solutions to the Maxwell equations. In this very limited sense there is a one-to-one correspondence between classical electromagnetic theory and quantum theory. The coherent states (which have now to be characterized by the entire set $\{\alpha_k\}$) are in one-to-one correspondence with the classical positive frequency solutions to the Maxwell equations. Of course, the way we construct a state of the field for all possible modes is to take the outer product, the direct product (which is what is meant by the product symbol Π_k) taken over all modes of the coherent states for each mode. Those are the coherent states, and some of their properties have been discussed in the chapter by Kibble.

The question now is, "Can we do something more with coherent states than simply say that this is a set of states which obeys our coherence conditions?" If that were all we could do with them, they would belong in a museum somewhere. The point is that they are very useful in doing calculations. First of all, it is useful to realize that any state of the field can be expressed in terms of the coherent states. Because there is such a vast number of coherent states, a two-dimensional continuum of them, and because they happen not to form an orthogonal set, there is a variety of ways in which we can expand any state, in terms of the coherent states. However, we are not going to expand states in terms of coherent states, we are not going to expand arbitrary state vectors, we are going to be concerned with operators (though the idea is basically the same). Suppose we want to expand any state in terms of the coherent states. We multiply by the unit operator which is $1/\pi$ times the dyadic product integrated over the entire complex plane, thus

$$|?\rangle = \frac{1}{\pi}\int |\alpha\rangle\langle\alpha|\,d^2\alpha\,|?\rangle, \tag{82}$$

where $d^2\alpha$ means the differential of the real part of α times the differential of the imaginary part of α.† Here we have explicitly an expansion of our unknown state in terms of coherent states. We can do that in general and we can expand operators in terms of the coherent states as well. The virtue of doing that, in particular for the density operator, is this. Once we have the density operator expressed in terms of the coherent states, then since everything we want to evaluate is in normally ordered form (at least for the moment) all of those operators applied to the coherent states on either side of them are turned into numbers and the operator aspect of the problem disappears. We then deal just

† $|?\rangle = \frac{1}{\pi}\int |\alpha\rangle\langle\alpha|?\rangle\, d^2\alpha$

$\langle\alpha|?\rangle = \exp(-\frac{1}{2}|\alpha|^2) f(\alpha^*)$ where $f(\alpha^*)$ is an entire function of α^*

with complex ordinary numbers. To expand the density operator, ρ, we multiply it by 1 on each side. Having used it twice, we have to integrate over two complex variables,

$$\rho = \frac{1}{\pi^2}\int |\alpha\rangle \langle\alpha|\rho|\beta\rangle \langle\beta| d^2\alpha\, d^2\beta. \tag{83}$$

Again, if we know the operator ρ, we know the function which stands in the middle of this expression. More generally for many modes, we have to expand in terms of a whole set of α-coefficients and a whole set of β-coefficients, as follows:

$$\rho = \frac{1}{\pi^2}\int\int |\{\alpha_k\}\rangle \langle\{\alpha_k\}|\rho|\{\beta_k\}\rangle \langle\{\beta_k\}| \prod_k d^2\alpha_k\, d^2\beta_k. \tag{84}$$

We do the same thing for every mode in the field. There are quite a few modes and quite a few integrals have to be done.

The point of this is that the expectation value of the field operators in a state like this (the matrix element between these states) reduces to a product of c-numbers.

$$\langle\{\beta_k\}| E^{(-)} \ldots E^{(+)} |\{\alpha_k\}\rangle = \Pi\mathscr{E}^*(\{\beta_k\})\, \Pi\mathscr{E}(\{\alpha_k\})\langle\{\beta_k\}|\{\alpha_k\}\rangle \tag{85}$$

All of the annihilation operators are applying to the right on a coherent state, so all of these annihilation operators become numbers, in other words this becomes just the product of eigenvalues $\mathscr{E}$. All of the creation operators are applying to the left on a coherent state, these turn into a product of complex conjugate eigenvalues. Then we have the integrations left which are implicit in this expression. The process of evaluating an expectation value becomes a process of integration over two complex variables for each mode. This is the most direct and the most obvious way of using the coherent states but there are other ways. The advantage of this particular procedure is that the sorts of functions that we deal with are always terribly well behaved. It is always true that the scalar product of the state α with anything can be expressed as $\exp(-\frac{1}{2}|\alpha|^2)$ times some function of the complex variable α^*, but it has to be an entire function. (An entire function is about the best behaved sort of function we can have — it has no singularities anywhere in the finite plane.) So those are non-singular functions, and, equivalently it is true in this representation that the weight function for the density operator, when multiplied by $\exp(\frac{1}{2}|\alpha|^2)\exp(\frac{1}{2}|\beta|^2)$ is likewise an entire function of α^* and β, again a very well behaved sort of function. We have that for the cost of doing two integrals for each mode.

There are other ways of proceeding. Let us first see something of the ambiguity in the use of the coherent states as an expansion basis. We can take the operator 1 and expand it in two ways in the coherent states, one way is this,

$$1 = \frac{1}{\pi^2}\int |\alpha\rangle \langle\alpha|\, 1\, |\beta\rangle \langle\beta| \mathrm{d}^2\alpha\, \mathrm{d}^2\beta$$

$$= \frac{1}{\pi^2}\int |\alpha\rangle \langle\beta| \exp(\alpha^*\beta - \tfrac{1}{2}|\alpha|^2 - \tfrac{1}{2}|\beta|^2)\, \mathrm{d}^2\alpha\, \mathrm{d}^2\beta. \qquad (86)\dagger$$

This is a representation of unity as a double integral in terms of the coherent state. But we know another representation, which is this,

$$1 = \frac{1}{\pi}\int |\alpha\rangle \langle\alpha| \mathrm{d}^2\alpha. \qquad (87)$$

If we do the β integration which is easy enough to do in this case, the first form of the representation degenerates into the second one. We can derive the second one from the first one. There are these two different ways of expanding this sample operator, unity, in terms of the dyadic product with different coherent state arguments and in terms of those with the same arguments, which is to say projection operators on the coherent states. These two varieties of expansion can be available for operators of all sorts and we shall next discuss the availability of the second kind of expansion for the density operator. We expand the density operator in the following way in terms of projection operators on the coherent states,

$$\rho = \int P(\alpha)\, |\alpha\rangle \langle\alpha|\, \mathrm{d}^2\alpha, \qquad (88)$$

where $P(\alpha)$ is a weight function which must be a real valued function of α in order to keep ρ Hermitian (as it must be). Is such a representation available? Well, thereby hangs a tale. Of course, if it does exist, then, since the trace of ρ is 1 and the trace of the dyadic product of the α state with itself is just unity, the function $P(\alpha)$ must be normalized when integrated over the entire complex plane,

$$\mathrm{Tr}\,\rho = 1 = \int P(\alpha)\, \mathrm{d}^2\alpha. \qquad (89)$$

If this representation exists, $P(\alpha)$ has some of the properties of a probability distribution. In fact, it is not one, and we shall see why below. But, is there such a representation as this and can we use it; and can we manage to survive what happens in the literature if we do?

† $\langle\alpha|1|\beta\rangle = \langle\alpha|\beta\rangle$
$\langle\alpha|\beta\rangle = \exp(\alpha^*\beta - \frac{1}{2}|\alpha|^2 - \frac{1}{2}|\beta|^2)$

7. THE P–REPRESENTATION

The point of expanding in terms of coherent states is that it simplifies the evaluation of statistical averages (expectation values). The expansion in terms of coherent states can be carried out in two ways and we illustrated both of those above. We recall that for the unit operator there are two species of expansion. The one is an expansion in terms of pairs of different coherent states and the other is an expansion in terms of pairs of identical coherent states. Now this second form of expansion is the simpler one since it involves only one complex variable for each harmonic oscillator in the field (only one complex amplitude for each of them). Let us talk therefore about *that* particular representation, which, wanting a better name, we call the "P-representation",

$$\rho = \int P(\alpha) |\alpha\rangle \langle\alpha| \, \mathrm{d}^2\alpha. \tag{90}$$

The function P should be real valued in order to keep ρ a Hermitian operator. There is a normalization condition on the density operator which is that the sum of its eigenvalues (which, as you know, represent probabilities) must be unity. The trace of ρ, therefore, is 1; that means in this representation that the integral of the function P taken over the entire complex plane has to be unity,

$$\int P(\alpha) \, \mathrm{d}^2\alpha = 1. \tag{91}$$

We have left completely open the question as to whether it is possible, in general, to construct such a representation. For some states obviously we can do it. For example, for a pure coherent state, the one based on the amplitude β,

$$\rho = |\beta\rangle \langle\beta|. \tag{92}$$

The way in which we do it is simply to choose for the function P, a δ-function,

$$P(\alpha) = \delta^{(2)}(\alpha - \beta), \tag{93}$$

where $\delta^{(2)}(\gamma)$ is a two dimensional δ-function. A 2-dimensional δ-function is the product of two one-dimensional δ-functions,

$$\delta^{(2)}(\gamma) \equiv \delta(\mathrm{Re}\,\gamma)\,\delta(\mathrm{Im}\,\gamma), \tag{94}$$

and the arguments of those functions are real, i.e. one is the real part of the argument, the other is the imaginary part of the argument; both of those are real numbers. The great advantage of this representation, where we can use it, is the following. If we have a normally ordered product of operators and want its statistical average, then we substitute for ρ the integral expression given in (90) for the density operator and the trace is then the average expectation value in a coherent state of the normally ordered product,

$$\mathrm{Tr}(\rho a^{\dagger n} a^m) = \int P(\alpha) \langle\alpha| a^{\dagger n} a^m |\alpha\rangle \, \mathrm{d}^2\alpha. \tag{95}$$

But now the operator a^m operates on a coherent state to the right and $a^{\dagger n}$ on a coherent state to the left; each of those operators then turns into its eigenvalue and what we are averaging is just a pair of complex numbers,

$$\mathrm{Tr}(\rho a^{\dagger n} a^m) = \int P(\alpha)\alpha^{*n}\alpha^m \, \mathrm{d}^2\alpha. \tag{96}$$

The averaging of operators turns into the averaging of numbers, but not only that, the averaging procedure looks a great deal like statistical averaging of the usual sort in probability theory.

There are a number of things to observe about this representation when we can use it. The temptation is very great to think of the function P as a probability density. All of the properties that we have indicated here are properties of a probability density. For example, when we have a perfectly definite state represented by a δ-function which is a non-negative function, or can be chosen as such, the averaging procedure we use is exactly what we would use in probabilistic terms. The only trouble is that this function P is *not* a probability density. It is *not* for many reasons which are all more or less related. The states that we label with the parameter α (the coherent states) are not an orthogonal set. So, for example, the projections in the directions of these coherent states in our entire space of states are not orthogonal projections. A harmonic oscillator can be in two different coherent states at once. We are not, therefore, summing mutually exclusive possibilities in summing these projection operators, so at least in that sense, P is not a probability density. It is not measurable experimentally as a probability density either, because α is an eigenvalue of a non-Hermitian operator, a, which has real and imaginary parts, or Hermitian and skew-Hermitian parts which do not commute and which are not, therefore, simultaneously measurable. They correspond to the position and momentum respectively of the oscillator and we cannot carry out a measurement which tells us anything definite about both position and momentum simultaneously. So there are no measurements we could make to determine a probability distribution (density) which would correspond to the function P. We have to think of the function P simply as a weight function in an expansion of this sort. Asymptotically, in the classical limit, it becomes a probability density. We shall see below a little more about what the classical limit is. In general, quantum mechanically, we have to face the possibility that the function P can take on negative values. (It is easy to find examples for which it does.) If P does happen to be a non-negative function it is easy to show that that is consistent with the positive definite character which the operator ρ must have, namely ρ as an operator can never have any negative eigenvalues. Indeed, positive functions P lead to *that* condition, but the converse is not true. Positive, definite and perfectly well behaved density operators do lead to functions P which freely take on negative values and there is in general, no avoiding that.

The function P is analogous to the probability density; asymptotically, in the classical limit, it is the probability density and it is probably best to give it the name, a name which was first used for another function, the name of quasi-probability density. The first such function which was known was that introduced by Wigner.[5] Wigner's interest was in finding the quantum mechanical analogue of the phase space density function in classical statistical mechanics. In classical statistical mechanics we are accustomed to talking about systems in terms of a density function in the space whose co-ordinates are position and momentum variables simultaneously. Now there is obviously something peculiar about doing that in quantum mechanics. It is only in a very limited sense that we can discuss such a function. It is obvious that no such thing can have the interpretation of a probability density, in general. Well, Wigner found such a function. It plays, as we shall see, for certain orderings of operators, a role analogous to that of the probability density. The particular operator orderings involved are ones which are symmetrical in the p and q variables. That happens to be the same statement as the statement that the orderings are symmetrical in the a and $a^\dagger$ variables. The Wigner function has a particular range of uses. It is most useful, naturally, when we are talking about symmetrically ordered variables but they are not what comes out as a rule in describing the simplest experiments in quantum optics. Well, our function P is not the Wigner function, it is another quasi-probability density, but, as we shall see below, a fairly close relative of the Wigner function.

Now, let us see what happens when we deal with a slightly more complicated situation in which many modes are excited. Then, of course, we have to deal with a many variable function $P(\{\alpha_k\})$. We deal with a function in which one amplitude is specified for every mode of the field. If we just define a symbol now which is the field eigenvalue corresponding to a particular coherent state, we see, if the coherent state is the one labelled with all of the amplitudes α_k, we can write the eigenvalue of the positive frequency part of the fields as $\mathscr{E}$ and it will be an expression which is linear in the amplitudes α_k,

$$E^{(+)}(r, t) \mid \{\alpha_k\}\rangle = \mathscr{E}(r, t\{\alpha_k\}) \mid \{\alpha_k\}\rangle. \tag{97}$$

The expression is a linear combination, an infinite sum, a sum over all the modes of terms which are linear in the amplitudes α_k. Now suppose we are told that such a P-representation exists for the density operator of the entire field, how do we calculate the correlation functions? Let us illustrate this for the first order correlation function. The story, of course, is the same for all of them. The first order correlation function is the statistical average of the normally ordered product of the field operators, and because it is a normally ordered product, we can just replace the field operators by their eigenvalues. The first order correlation function expressed as an integral taken over all of

the complex mode amplitudes α_k is given by

$$G^{(1)}(X_1, X_2) = \int P(\{\alpha_k\})\, \mathscr{E}^*(X_1\{\alpha_k\})\, \mathscr{E}(X_2\{\alpha_k\})\, \Pi\, \mathrm{d}^2\alpha_k. \tag{98}$$

This is an exact quantum mechanical expression, and yet we see here something which looks a great deal like the classical procedure which we encountered in eqn. (12) for evaluating the classical first order correlation function. The correspondence is as follows:

$$\mathscr{E}(X\{\alpha_k\}) \longleftrightarrow E^{(+)}_{\text{class}}(X\{\mathscr{C}_k\}) \tag{99}$$

$$P(\{\alpha_k\}) \longleftrightarrow \mathscr{P}(\{\mathscr{C}_k\}) \tag{100}$$

$$i(\hbar\omega_k/2)^{\frac{1}{2}}\alpha_k \longleftrightarrow \mathscr{C}_k \tag{101}$$

The field eigenvalue $\mathscr{E}$ corresponds simply to the positive frequency part of the classical field. The weight function in our representation of the density operator, $P(\alpha)$, corresponds to the probability density for the random Fourier coefficients $\mathscr{C}_k$. Finally, the $\mathscr{C}$'s themselves correspond to particular given constants multiplying the amplitudes α_k. When we make these identifications, there is a precise correspondence between the formulae for the classical correlation function and the quantum mechanical one. Presumably, we use the classical procedure when the numbers are meant to describe strong fields. But, of course, if we are dealing with really strong fields, the number $\mathscr{C}_k$ is not an infinitesimal amplitude, it is substantial in typical classical units. Then since Planck's constant is so terribly small, that is going to mean that $|\alpha|$ is going to have to be very large, and so the classical limit, strictly speaking is the limit in which the dimensionless excitation moduli, $|\alpha|$, become very much larger than unity.

An illustration of the transition from the quantum to the classical limit may be seen in the analysis of the parametric amplifier by Mollow and Glauber.[6] The dynamical behaviour of an amplifier is discussed. Initially any field is put into the amplifier—any field at all. Now, in general, for arbitrary quantum fields, the function P can take on negative values of course, but worse still, it can be quite singular; we see one example of it in the case of the coherent state itself in which P is a δ-function. But, as we shall see in a little more detail below, the function P can be even more singular than a δ-function, and in fact it can be so singular that there is a serious question whether it exists at all. We put such fields (for example for arbitrarily singular functions P) into our model amplifier and let the amplifier amplify and develop the function P as a function of time. The density operator of the field in the amplifier of course keeps changing as the amplifier amplifies it and what we discover is that, physically, the amplifier turns this, perhaps poorly behaved function P at time zero, into an exceedingly smooth and well-behaved function at large time. If P took on negative values at time zero, it turns out there is a time beyond which it can

only take on positive values. The function P depends, in other words, on the initial field. The field in the amplifier is described by a function P which eventually becomes very smoothly behaved over distances in the complex plane of the order of one unit and takes on all of the properties of a probability density in the classical limit in which it really becomes one.

We have so far provided a precise, non-trivial justification for the procedure which has always been used in optical calculations, where it has been used correctly. The grand tradition in doing optical calculations in determining the intensities of interference patterns and the like, has been to assume that a classical field is incident on the system, to calculate the field amplitude where it is being observed and then to average that field amplitude over some sort of distribution. Typically, we do not deal with the absolute intensity anyway; we normalize it relative to the input intensity. In that case, we are carrying out exactly this averaging procedure and we have shown that for the class of experiments in which we are detecting what amounts to the averages of normally ordered operators, that is a precisely correct procedure to carry out. The general characterisation of such experiments is that they are the ones which give null results in the vacuum. It is not really necessary to call that the semi-classical method, or quasi-classical, or anything of the sort, it is precisely quantum mechanical provided we know that the representation of the field we are dealing with exists in this particular form in terms of the coherent states (P-representation). Now, of course, if we are doing a different experiment, which is based upon a different ordering of the operators, then the story is different. There is no statement about the strength of the field required in order to use this particular representation, it holds for arbitrarily weak fields. In that sense, it is a completely quantum mechanical result—we simply should not describe it as classical in any sense at all.

Now we have to face the question "Can we find such a representation?" and, "How, in particular, do we discover what the function P is for any given field?" There is a number of such questions which we can gather together and discuss as a unit. First let us note that there is a whole class, a whole family of functions which we ought to identify as quasi-probability densities. One of them is the function P, above, another one is the Wigner function which is now some 38-39-years-old, and still another is a function which we can form very easily by taking the diagonal matrix element of the density operator in a pure coherent state; that, obviously, is something which is non-negative, it is greater than or equal to zero, because the density operator is a positive operator,

$$P(\alpha), \quad W(\alpha), \quad \langle\alpha|\rho|\alpha\rangle \geqslant 0. \tag{102}$$

The functions (102) are quasi-probability densities because again there is no way in which we could measure them having measured the real and imaginary

parts of the co-ordinate a and observing the frequencies with which they occur (notwithstanding the fact, in other words, that $\langle\alpha|\rho|\alpha\rangle$ is a positive function, it is still a quasi-probability density). These are three particular examples of a more general family of quasi-probability densities, $W(\alpha, s)$, distinguished by the parameter s. This general family of quasi-probability densities is described in detail by Cahill and Glauber.[9] There are many mathematical theorems we can prove about them and we shall only discuss one or two of them.

One theorem which is very convenient to know about, if we want to order creation and annihilation operators, is the theorem for multiplying together two exponential functions of operators which have a rather simple commutation relation. We assume that their commutator commutes with each of them. Obviously this is true for creation and annihilation operators because the commutator is unity or zero. The product of two such exponentials is not simply $\exp(A+B)$ because A and B do not commute but all the correction we need is to add in the exponent the quantity $\frac{1}{2}[A, B]$, which can be treated as a c-number since it commutes with both A and B,

$$\exp A \exp B = \exp(A+B+\tfrac{1}{2}[A, B]). \tag{103}$$

This holds for all operators A and B such that

$$[A[A, B]] = [B[A, B]] = 0. \tag{104}$$

The simplest way to prove this theorem is just to put a parameter, say λ, in the exponent, differentiate with respect to the parameter and then observe that the differential equation obtained may be immediately integrated to give the result. The proof is in Messiah's book on quantum mechanics.[7]

Let us now define a unitary displacement operator $D(\lambda)$. There is in the exponent a skew-Hermitian operator. This is an operator analogue of, for example, $\exp[i(\text{real number})]$. It is a unitary operator in the sense that $D^{-1} = D(-\lambda) = D^{\dagger}(\lambda)$. Now, when we consider ordinary probability densities, it is useful, frequently to define the Fourier transform of the probability density. There are a great many calculations done in probability theory which are done more easily that way; it is the so-called characteristic function. However, in considering a density operator in the present case we have to find a somewhat different analogue for the construction of the characteristic function. We shall still call what we construct a "characteristic function" but we shall take it to be formed in this particular way,

$$X(\lambda) = \mathrm{Tr}\{\rho D(\lambda)\} = \mathrm{Tr}\{\rho \exp(\lambda a^{\dagger} - \lambda^* a)\} \tag{105}$$

where the Fourier transform of the probability density is really the expectation value of the Fourier exponential function. Here we simply take that expectation value by taking the trace of the density operator times the unitary operator D.

It is implicit in this exponential function that operators a and $a^\dagger$ are treated in similar ways and if we were to expand this exponential function of the operators we would find that the operators are ordered in all possible ways in each term of the expansion. This, in other words, is a symmetrically ordered expression when we expand it.

Now, there are other characteristic functions that we can define in which the operators would not be ordered exactly symmetrically. For example, we can define another version of the characteristic function in which we separate the exponential functions of the two operators (that is of course simply a different function). Here the operator content of the expectation value is written in normally ordered form:

$$\mathrm{X}_N(\lambda) = \mathrm{Tr}\{\rho \exp(\lambda a^\dagger) \exp(-\lambda^* a)\}. \tag{106}$$

Of course, that has to be related to the characteristic function defined by (105) and, using the multiplication theorem for exponentials, we obtain the latter with the correction factor $\exp(\frac{1}{2}|\lambda|^2)$,

$$\mathrm{X}_N(\lambda) = \mathrm{Tr}\{\rho \exp(\lambda a^\dagger - \lambda^* a)\} \exp(\tfrac{1}{2}|\lambda|^2). \tag{107}$$

We could equally well define a characteristic function in which the operators are in antinormal form and the correction factor is $\exp(-\frac{1}{2}|\lambda|^2)$,

$$\mathrm{X}_A(\lambda) = \mathrm{Tr}\{\rho \exp(-\lambda^* a) \exp(\lambda a^\dagger)\} \tag{108}$$

$$= \mathrm{Tr}\{\rho \exp(\lambda a^\dagger - \lambda^* a)\} \exp(-\tfrac{1}{2}|\lambda|^2). \tag{109}$$

We are considering here two examples of something which is obviously more general.

We can define a characteristic function which depends not only on the arbitrary parameter λ but on a new variable s; we shall define it with a correction factor $\exp(\frac{1}{2}s|\lambda|^2)$ thus,

$$\mathrm{X}(\lambda, s) = \mathrm{X}(\lambda) \exp(\tfrac{1}{2}s|\lambda|^2). \tag{110}$$

Then, having done that, we can identify the three characteristic functions which correspond to different values of the parameter s. When $s = 1$, we have the normally ordered one, when $s = 1$ we have the anti-normally ordered one, when $s = 0$ we just have the one defined originally,

$$\begin{aligned} \mathrm{X}(\lambda, 1) &\equiv \mathrm{X}_N(\lambda), \\ \mathrm{X}(\lambda, 0) &\equiv \mathrm{X}(\lambda), \\ \mathrm{X}(\lambda, -1) &\equiv \mathrm{X}_A(\lambda). \end{aligned} \tag{111}$$

We can use the exponential function to define s-ordered products of operators,

$$\exp(\lambda a^\dagger - \lambda^* a + \tfrac{1}{2}s|\lambda|^2) = \sum \frac{\lambda^n(-\lambda^*)^m}{n!\,m!}\{a^{\dagger n}a^m\}_s. \tag{112}$$

This is very convenient for the purposes of dealing with all orderings at once. Suppose we write the exponential operator and add a correction factor. When $s = 1$ we know that this exponential is identically equal to the normally ordered exponential. When $s = -1$ we know that this exponential is identically equal to the anti-normally ordered exponential function. When $s = 0$ it is obviously the symmetrical one. Those three cases, in other words, are included and then, of course, by letting the parameter s take on other values, we include many other "orderings". In a formal sense they are orderings of operators but I do not know how we would write them if, for example $s = \sqrt{1/\pi}$; it is simply a formal notion, but it does allow us to deal with the above three orderings together, and to see something which we would not otherwise see of the relationship among the functions (the auxiliary functions) for each variety of operator ordering. The right-hand side of (112) is obtained by expanding the exponential function. We can use the terms of the power series expansion to define s-ordered operators and then for $s = 1$ we are dealing with normally ordered combinations (normally ordered products), for $s = -1$ we are dealing with anti-normally ordered ones, and for $s = 0$, this is a definition of symmetrically ordered ones (and what we mean by symmetrically ordered there, is really taking the arithmetic mean of all the possible ways of arranging these particular factors in each term of the expansion).

Now we can see the following things. If we want to evaluate the statistical average of such an s-ordered product, we can do that by going back to the appropriate characteristic function, differentiating with respect to the parameter λ, or λ^* an appropriate number of times, and then setting $\lambda = 0$,

$$\frac{\partial^n}{\partial\lambda^n}\,\frac{\partial^m}{\partial(-\lambda^*)^m}\,\mathrm{X}(\lambda, s)\Big|_{\lambda=0} = \mathrm{Tr}[\rho\{a^{\dagger n}a^m\}_s]. \tag{113}$$

This s-ordered characteristic function we have defined is really a generating function for the statistical averages of the arbitrarily ordered products of operators. If we know the characteristic function, then we find the averages just by evaluating all of its derivatives at the origin, at $\lambda = 0$.

Now let us look at the case which interests us most, the case of normal order, the normally ordered characteristic function, for which $s = 1$. Let us assume that P-representation exists and, in that case, we evaluate that characteristic function by just taking the statistical average of the normally ordered exponential function, and that is a trivial thing to do. Since the exponential is normally ordered, each of the operators in it simply becomes an ordinary

number multiplying a coherent state; the scalar product of the two coherent states is unity and they go away and we are left then with this explicit evaluation of the normally ordered characteristic function,

$$\mathrm{X}(\lambda, 1) = \mathrm{X}_N(\lambda) = \int P(\alpha) \langle \alpha | \exp(\lambda a^\dagger) \exp(-\lambda^* a) | \alpha \rangle \, \mathrm{d}^2\alpha$$
$$= \int \exp(\lambda\alpha^* - \lambda^*\alpha) P(\alpha) \, \mathrm{d}^2\alpha. \tag{114}$$

The exponential function here has an imaginary argument; it is a function of the two complex variables λ and α and from an algebraic standpoint, it is simplest at this stage to use Cartesian coordinates. When we do that we see that we have a Fourier transform in two variables of the function P. The Fourier transform is, presumably, invertible (that is a bit of a question here, we will have to come back to it). Let us assume that the Fourier transform *is* invertible. A convenient way in which to write the Fourier integral here uses the Dirac–Fourier integral representation of the δ-function written out in two dimensions and in complex notation. This particular form of integral representation is,

$$\frac{1}{\pi^2} \int \exp[\lambda(\alpha^* - \beta^*) - \lambda^*(\alpha - \beta)] \, \mathrm{d}^2\lambda = \delta^{(2)}(\alpha - \beta). \tag{115}$$

By multiplying eqn. (114) by an exponential function of λ of the appropriate form and integrating, we, in effect, project out the function P,

$$P(\alpha) = \frac{1}{\pi^2} \int \mathrm{X}_N(\lambda) \exp(\alpha\lambda^* - \alpha^*\lambda) \, \mathrm{d}^2\lambda. \tag{116}$$

The function P is thus the corresponding Fourier integral of the normally ordered characteristic function, providing we can invert the Fourier transform.

The problem in inverting the Fourier transform is that for some states (and these are perfectly reasonable ones,) the normally ordered characteristic function, though it is a continuous and very well-behaved function, becomes extremely large as the modulus of λ goes to infinity. The only general bound that we can put on the normally ordered characteristic function comes from this consideration: The ordinary characteristic function, the one which is the expectation value of the unitary operator, has to be bounded by unity, simply because it is the average of a unitary operator,

$$|\mathrm{X}(\lambda)| \leqslant 1. \tag{117}$$

However, since the normally ordered characteristic function is equal to the ordinary one times a Gaussian factor, then we see that the normally ordered

characteristic function is only bounded by a function which blows up as λ goes to infinity,

$$|X_N(\lambda)| \leqslant \exp(\tfrac{1}{2}|\lambda|^2). \tag{118}$$

Now, we can indeed find quantum states, which at least come close to meeting this bound as $\lambda \to \infty$ and then we are in a terrible position; if we insist upon inverting the Fourier transform (114), we have to calculate the Fourier integral of a function which blows up as $\exp(+x^2)$, as it were. We can find examples, and many of them, for which that happens in at least one of the two variables of integration. In other words, there are cases in which it does not seem possible to define the function P in any familiar sense in Fourier transform theory. Certainly not in the sense of square integrable functions, and in fact not more generally in the much broader context of tempered distributions which is discussed these days in connection with Fourier transform theory.

Now, we can take either of two attitudes towards this problem which seem to have evolved in the literature. One attitude is that when we are dealing with quantum states for which the function becomes so enormously singular, there is really not much convenience left in defining the P-representation or attempting to use it, and there is certainly very little sense in saying that we have any remaining resemblance between the quantum mechanical and classical pictures. A different attitude is that we can define, perhaps not a P-representation in the sense that we have been describing, but we can, for example, do calculations by using limiting procedures by representing the density operator as a limit of an approximating sequence of P-representations. That has been the attitude of Klauder and Sudarshan.[8] Their coworkers have gone on to identify integrals of this sort, not with distributions or tempered distributions in the ordinary sense, but with things which are now being called ultra-distributions. They are still a vastly more general notion of a generalized function. This is rather deep mathematical water because once we have defined integrals of this sort, it is not at all clear what we do with them. Thus far, nothing more than the definition seems to exist.

There does not seem to be a clear or simple answer to the question of whether the P-representation can be constructed in general. It depends, at least in part, on definition. Now, for the other quasi-probability densities, the situation tends to be rather simpler and we can deal with them all at once in the following way.

We can define a function $W(\alpha, s)$, where s is the ordering parameter, and take that in each case to be the Fourier transform of the appropriately ordered characteristic function,

$$W(\alpha, s) \equiv \int X(\lambda, s) \exp(\alpha\lambda^* - \alpha^*\lambda) \frac{d^2\lambda}{\pi}. \tag{119}$$

Then the formula (114) is precisely this for the case $s = 1$ except that we have put a factor of π into the formula for W,

$$W(\alpha, 1) = \pi P(\alpha).$$

We can invert the transform (119), in general, (or let us assume that we can); then the ordered characteristic function is written as a Fourier integral

$$\mathrm{X}(\lambda, s) = \int \exp(\lambda\alpha^* - \lambda^*\alpha)\, W(\alpha, s) \frac{\mathrm{d}^2\alpha}{\pi}. \tag{120}$$

Suppose we differentiate the inverted transform (120) in the neighbourhood of the origin. We have already shown that is the statistical average of the s-ordered product of operators (113). Now it is obvious that if we differentiate the exponential in (120) and then set $\lambda = 0$, we are finding the moments of the function W,

$$\frac{\partial^n}{\partial\lambda^n} \frac{\partial^m}{\partial(-\lambda^*)^m} X(\lambda, s)\Big|_{\lambda=0} = \mathrm{Tr}[\rho\{a^{\dagger n} a^m\}_s] = \int \alpha^{*n}\alpha^m W(\alpha, s) \frac{\mathrm{d}^2\alpha}{\pi}. \tag{121}$$

Now for all of these orderings, for the three familiar orderings and for all of those that we define formally for all complex values of s, the following is true. The statistical average of the ordered product of operators is given by the ordinary c-number moments, the classical moments, so-to-speak, of the function W. In particular, for $n = m = 0$ we have just the normalization relation,

$$\int W(\alpha, s) \frac{\mathrm{d}^2\alpha}{\pi} = 1, \tag{122}$$

because, of course, the characteristic function at argument zero is just the trace of the density operator, that is, just unity. So (122) is a normalization condition for the entire family of quasi-probability distributions. For $s = 0$, we have precisely the function W which was defined by Wigner in terms of real variables, not in terms of complex amplitudes, many years ago. For $s = -1$ we have anti-normal order ($\mathrm{X}(\lambda, -1)$ is the anti-normal characteristic function), that is, just the average of the pair of exponential functions which are written in anti-normal order

$$\begin{aligned} \mathrm{X}(\lambda, -1) \equiv \mathrm{X}_A(\lambda) &= \mathrm{Tr}\{\rho \exp(-\lambda^* a) \exp(\lambda a^\dagger)\} \\ &= \mathrm{Tr}\left\{\rho \exp(-\lambda^* a) \frac{1}{\pi}\int |\alpha\rangle\langle\alpha|\, \mathrm{d}^2\alpha \exp(\lambda a^\dagger)\right\}. \end{aligned} \tag{123}$$

Here we have written that same statistical average on the line beneath but we have written the operator 1 in between the two exponential functions as an expansion in terms of the coherent states. Now we evaluate the trace. To

evaluate the trace, of course, we push the integral sign to one side and the differential to the other side with $(1/\pi)$. To evaluate the trace we just take the expectation value of the expression inside the brackets due to the coherent state. But $\exp(\lambda a^\dagger)$ is applied from the right to the left on a coherent state, the exponential function $\exp(-\lambda^* a)$ is applied to a coherent state, so those turn into ordinary numbers and what remains finally is just an integral, in fact a Fourier integral of the diagonal matrix element of the density operator ρ,

$$\mathrm{X}(\lambda, -1) = \int \exp(\lambda\alpha^* - \lambda^*\alpha) \langle\alpha|\rho|\alpha\rangle \frac{\mathrm{d}^2\alpha}{\pi}. \tag{124}$$

So, in other words, the anti-normally ordered characteristic function is just the Fourier transform of the diagonal matrix element of ρ.

Now we defined the function W for the general case of arbitrary s as the Fourier transform of X, and X is consequently the Fourier transform of W. Well, we can conclude from this, that $W(\alpha, -1)$ is just the diagonal matrix element of the density operator in a coherent state,

$$W(\alpha, -1) = \langle\alpha|\rho|\alpha\rangle. \tag{125}$$

There are many properties of the Wigner function that we easily prove this way but we are not too interested in symmetrical ordering so let us pass over them. We have given three identifications of values of the function W. The first is that $W(\alpha, +1)$ is πP, the second is that $W(\alpha, 0)$ is just the Wigner function, and finally, $W(\alpha, -1)$ is simply the diagonal element of the density operator in a coherent state.

An example of the sort of relationship between these functions may be proved easily in the following way. The ordered characteristic function we have defined is just the ordinary one times $\exp(\frac{1}{2}s|\lambda|^2)$. So the relationship between two of these ordered characteristic functions is obviously given by

$$\begin{aligned} \mathrm{X}(\lambda, s) &= \mathrm{X}(\lambda) \exp[(\tfrac{1}{2}s|\lambda|^2)], \\ \mathrm{X}(\lambda, s) &= \mathrm{X}(\lambda, t) \exp[\tfrac{1}{2}(s-t)\,|\lambda|^2]. \end{aligned} \tag{126}$$

The quasi-probability functions W are the Fourier transforms of the characteristic functions. We take the Fourier transform of both sides of (126). On the right we must have a convolution integral, and the kernel of that convolution integral is just the Fourier transform of this particular Gaussian function. That means that we have the following general relation among these quasi-probability densities,

$$W(\alpha, s) = \frac{2}{t-s} \int \exp[-2\,|\alpha-\beta|^2/(t-s)]\, W(\beta, t) \frac{\mathrm{d}^2\beta}{\pi},$$

for $$\mathrm{Re}\, t > \mathrm{Re}\, s. \tag{127}$$

The $W(\alpha, s)$ are all related to one another, they are all Gaussian convolution integrals of one another. For example, if $t = 1$ and $s = -1$, the diagonal matrix element of the density operator is just this particular integral,

$$\langle\alpha|\rho|\alpha\rangle = \int \exp(-|\alpha-\beta|^2) P(\beta)\, d^2\beta. \tag{128}$$

This is a trivial relationship, it is one which we see immediately by just taking the expectation value of the P-representation.

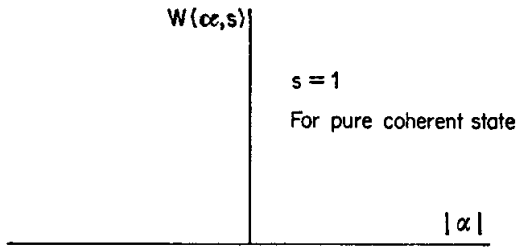


FIG. 6

There is a large family of such relationships. What happens, in fact is this. As we go along the succession of orderings from $s = 1$ to $s = -1$, we find smoother and smoother functions. A state, let us say a pure coherent state, for which we have a delta function corresponding to normal ordering, corresponds to a Gaussian function for $s = -1$, see Figs. 6 and 7. There is a tendency always to smooth the functions as s decreases. For a more general state, the function P might be something which wiggles a great deal. Figure 8 is an example with a good deal of wiggle in it, including negative values. As we go towards the other end of our spectrum of quasi-probability functions, and go eventually toward those which deal with anti-normally ordered operators we tend to find smoother versions of the functions, for example, we

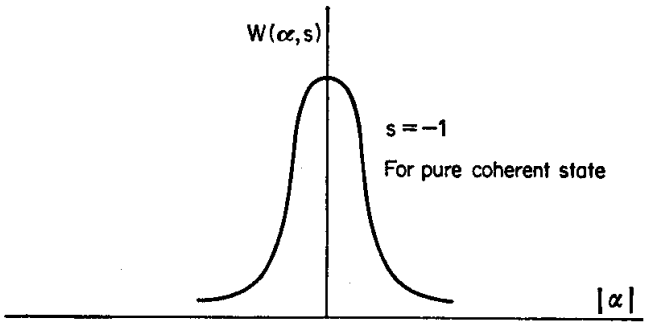


FIG. 7

discover that the Gaussian function which carries out the smoothing process wipes out the negative values entirely and we have a function which is never negative as shown in Fig. 9.

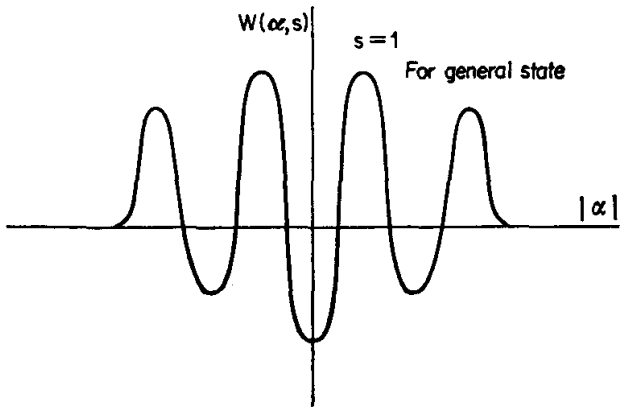


FIG. 8

There are many games we can play with these functions; we can answer a great many questions about how explicitly to order operators. For example, we might imagine that, given any particular operator, then it can easily be cast into anti-normally ordered form, or normally ordered form. If we are dealing with a polynomial in the creation and annihilation operators, it is obvious that that can be done by just using the commutation relations. But it is much less obvious whether those two operations can be carried out when we are dealing with infinite series in the creation and annihilation operators. In this case, the peculiar result that emerges is this: A very broad class of operators can, indeed, be expanded in terms of normally ordered products, but it is only a very much smaller class of operators, which can be expanded in terms of anti-normally ordered products of creation and annihilation operators. That is simply a mathematical result. It is one which again stands in the way of the general construction of the *P*-representation. One of the advantages of this generalized treatment of orderings is that we have the parameter *s* with which we can attempt to define the *P*-representation in singular

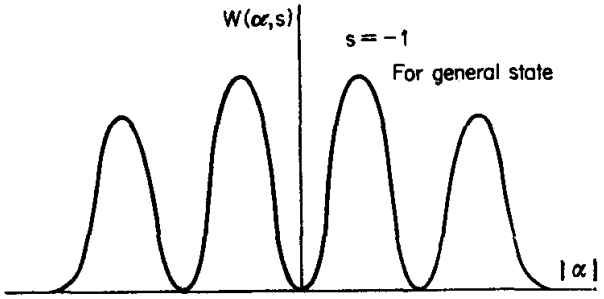


FIG. 9

examples. It turns out that that procedure actually fails, that there is no means by which we can define the P-representation in general, for all possible states. At least, there is no method within this formalism which permits us to do this.

These questions of operator ordering, expansions, and the like are not physics. They are, at best, a matter of mathematical convenience, and at worst, a kind of preoccupation which consumes us as we discover more and more interesting theorems. Let us get back to physics.

Unhappily, the most typical state of the world is not a pure coherent state. We have lived for a long time without having to pay any attention to coherent states, physically. The most typical state of the world, the one which seems to prevail in the overwhelming proportion of cases, is the state of chaos. To define chaos we define the entropy of the field. The entropy of any chaotic system has this familiar definition which goes back to von Neumann,

$$S = -\mathrm{Tr}\,(\rho \log \rho)_{\max}\,. \tag{129}$$

There are some constraints in maximizing the entropy. Obviously, the trace of the density operators has to remain 1. Suppose we fix the mean number of quanta present in the field, we then have

$$\mathrm{Tr}\,\rho a^{\dagger}a = \langle n\rangle. \tag{130}$$

The problem then is to maximize (129) with the constraint of (130). In saying that the typical state of the world is chaos, all we are saying is what any statistical mechanician would say (they are all cynics). We have to quote some good reason to have the world in a state other than one of chaos. There are quite frequently good reasons. But if someone gives us a light source and asks for our best guess, of its state, saying that it has such and such an intensity, then the very best guess, in the absence of information is the one which maximizes entropy. That is the statement of information theory. It is very easy to carry out the mathematical operation using Lagrange multipliers to maximize the entropy and the density operator that we get for a single mode is this:

$$\rho = \frac{1}{1+\langle n\rangle}\left\{\frac{\langle n\rangle}{1+\langle n\rangle}\right\}^{a^{\dagger}a}. \tag{131}$$

That may not look like a familiar density operator, but in fact we are quite familiar with it for the case of thermal excitation, which, by the way, is only one particular example of chaos. (It is really making a mistake to say that all chaos is thermal, the radiation which emerges from an electric discharge tube at an energy of a few electron volts is certainly chaotic in character, but it is by no means thermal.) For the thermal case

$$\langle n\rangle = \frac{1}{\exp\left[(\hbar\omega/kT)-1\right]}.$$

Then (131), in the thermal case becomes the Planck density operator.

It is easy to evaluate the functions (102). To obtain the diagonal matrix element of the density operator, we just sandwich ρ between two coherent states,

$$\langle\alpha|\rho|\alpha\rangle = \frac{1}{1+\langle n\rangle}\exp(-|\alpha|^2)\sum_j \frac{|\alpha|^{2j}}{j!}\left(\frac{\langle n\rangle}{1+\langle n\rangle}\right)^j. \tag{132}$$

We can easily evaluate the infinite sum and get

$$\langle\alpha|\rho|\alpha\rangle = \frac{1}{1+\langle n\rangle}\exp\left[-|\alpha|^2/(1+\langle n\rangle)\right] = W(\alpha,-1). \tag{132}$$

Taking the Fourier transform, we obtain the characteristic function for $s=-1$,

$$\mathrm{X}(\lambda,-1) = \exp\left[-(1+\langle n\rangle)|\lambda|^2\right]. \tag{134}$$

If we then multiply by the exponential function which gives us the general form of the characteristic function for all values of the ordering parameter, we have the result:

$$\mathrm{X}(\lambda,s) = \exp\left[-\left(\frac{1-s}{2}+\langle n\rangle\right)|\lambda|^2\right]. \tag{135}$$

Now, if we go back and take the Fourier transform once more we have the result finally, for the whole family of quasi-probability distributions,

$$W(\alpha,s) = \frac{2}{2\langle n\rangle+1-s}\exp\left(\frac{-2|\alpha|^2}{2\langle n\rangle+1-s}\right). \tag{136}$$

In particular, for $s=1$, we have the simple Gaussian function,

$$P(\alpha) = \frac{1}{\pi}W(\alpha,1) = \frac{1}{\pi\langle n\rangle}\exp\left(-|\alpha|^2/\langle n\rangle\right). \tag{137}$$

Nothing could be more obvious or simple for the case of chaos. One of the best ways, perhaps, of defining chaos, is to say it is what we must have when we have many independently contributing elements. If we take any light source, which is divisible in principle, into many small parts, and assume that they all contribute independently, then it is inevitable from the central limit theorem of probability theory (there is a simple generalization in the quantum mechanical case), that we have a Gaussian distribution of amplitudes. That is precisely what we mean by chaos. We shall see below what results from this chaos as far as measurements on the field are concerned.

8. Chaotic States

All of the older and more traditional measurements in optics are ones that we can describe in terms of the first order correlation function $G^{(1)}$. In a sense, the older optics by measuring intensities only, really looks no further into the field than into the contents of the function $G^{(1)}$. For the case of a stationary field, as we shall see, the first order correlation function is determined ordinarily just by the spectral data of the field, i.e. the intensities of excitation of the various modes of the field. When we say spectral data we mean polarization data as well as the usual sort of spectral data. We can imagine different sorts of fields, which have identical spectra. We might take them from different sources, such as a laser or a black-body; we might have intensity problems, but we can, presumably, filter black body radiation to achieve any spectral radiation distribution that we like, including the distribution which comes from a laser. Then we might ask the question, "Are these two fields distinguishable?" If their spectra are identical, the functions $G^{(1)}$ will be identical, and then no matter which of the traditional optical experiments that we perform, we shall never be able to distinguish between these two light beams.

On the other hand, the distinction becomes very clear when we begin to look at the effects that are involved in the higher order correlation functions. The first of these was the HBT effect. We have to measure the second order correlation function or some higher order correlation function in order to see the difference. For the particular case of chaotic fields, it must still be true that these higher order correlation functions are determined simply by the spectral data of the field. We know that for a multi-mode, chaotic state, the field (P-representation) can be described in terms of a multivariate Gaussian function

$$P(\{\alpha_k\}) = \prod_k \frac{1}{\pi \langle n_k \rangle} \exp\left(-|\alpha_k|^2/\langle n_k \rangle\right). \tag{138}$$

The only information that we need in order to construct this function is just the mean number of photons in each mode. Those are exactly the same data that are contained in the first order correlation function. So, for the case of a stationary Gaussian field, i.e. a chaotic state, the entire density operator contains no more information than is contained in just the first order correlation function. If we measure the first order correlation function for such fields, we measure everything. Now that is not to say that the higher order correlation functions have no interesting structure, in fact they have some structure, but it must be possible for the case of chaotic fields to construct the higher order correlation functions from a knowledge simply of the first order correlation

function. There is a theorem which permits us to do that. It is a generalization of a theorem which holds in the theory of stochastic processes quite generally. If we use this Gaussian distribution (138) to construct the correlation function of nth order, we find (remember it has altogether $2n$ different arguments, in general) that the higher order correlation function can always be expressed as the sum of products of the first order correlation functions and for the nth order correlation function we have to form an n-fold product; we have to sum n-fold products over all possible permutations of the coordinates,

$$G^{(n)}(x_1 \ldots x_n y_1 \ldots y_n) = \sum_P \prod_{j=1}^{n} G^{(1)}(x_j y_{P_j}). \tag{139}$$

In other words, if the first coordinate in $G^{(1)}$ takes on all the values from $1 \to n$, the coordinates associated in the same functions with those coordinates constitute the $n!$ permutations of the set of numbers $1, \ldots, n$.

As an example of such a function, consider the two-fold joint counting rates, the second order correlation function, in other words, with its arguments repeated; x_1 occurs twice, x_2 occurs twice. That is a sum of two-fold products of correlation functions. The identity permutation corresponds to the first term, and then the simple interchange of the x_1 and x_2 coordinates determines the second term. The two factors are complex conjugates of one another so we simply write the squared absolute value, for the second term:

$$G^{(2)}(x_1 x_2 x_2 x_1) = G^{(1)}(x_1 x_1)\, G^{(1)}(x_2 x_2) + |G^{(1)}(x_1 x_2)|^2. \tag{140}$$

If the field were a coherent one, only the first term would be present, the correlation function would factorize and all we would have is

$$G^{(2)}(x_1 x_2 x_2 x_1) = G^{(1)}(x_1 x_1)\, G^{(2)}(x_2 x_2).$$

This means that there would be no correlation at all in the counting rates observed by two counters placed in the field; there would be no tendency towards photon coincidences. The additional term $|G^{(1)}(x_1, x_2)|^2$ is due to the fact that the field to be averaged has a Gaussian distribution of field amplitudes. The term $|G^{(1)}(x_1, x_2)|^2$ is the one responsible for the HBT effect.

Let us now see how we calculate first order correlation functions. In most of the stationary fields we work with, we can always choose a suitable set of modes such that the mean value of the products of the creation operator for the mode k and the annihilation operator for the mode k' vanishes unless the modes k and k' are the same and then, of course, it must be just the mean number of quanta in the mode,

$$\mathrm{Tr}\,\{\rho a_k^{\dagger} a_{k'}\} = \langle n_k \rangle \delta_{kk'}. \tag{141}$$

Now in the stationary field, the expectation value when these mode indices are different must vanish if the modes are at different frequencies. If the modes

are of the same frequency, it is possible to have non-vanishing values for different modes, but then a suitable choice of the modes k and k' (in other words the use of the correct principal axes in the Hilbert space of modes) will lead to a reduction of the expectation value to the form (141) in any case. Now if this is true, then we find the first order correlation function by just substituting for the expectation values in the formula for the expansion of $G^{(1)}$ in terms of the mode amplitudes, and we obtain

$$G^{(1)}(rt, r't') = \tfrac{1}{2}\sum_k \hbar\omega_k \langle n_k\rangle u_k^*(r)\, u_k(r') \exp[i\omega_k(t-t')]. \tag{142}$$

We see how for the stationary field simply the set of spectral data (the set of mean occupation numbers) determines the first order correlation function.

Consider a specific example, say a beam of plane waves. As long as we have the symmetry which describes plane waves, i.e. a beam moving uniformly in the $+x$ direction for which the mode function is $u_k(x) \sim \exp(ikx)$, there need be only one argument in the first order correlation function. We shall call that argument τ,

$$G^{(1)} = G^{(1)}(\tau), \qquad \tau = t_1 - t_2 - \frac{1}{c}(x_1 - x_2). \tag{143}$$

The function $G^{(1)}$ is independent of y and z if x is the direction of propagation of the waves. Let us make a spectral assumption about the plane waves; a simple one to make is that the occupation numbers have a Gaussian behaviour, i.e. we have a Gaussian line profile such as may be given by Doppler broadening.

$$\langle n_\omega\rangle = \text{constant} \cdot \exp[-(\omega-\omega_0)^2/2\gamma^2]. \tag{144}$$

We could work with any line profile but since we have to take the Fourier integral of the spectral density, the choice of a Gaussian makes this easy. The integral is the summation over modes. We assume that the modes are closely spaced and therefore we can integrate over the propagation vectors or, equally well, frequencies (since we have specified the direction of propagation). We find the first order correlation function in terms of the interval variable τ is given by

$$G^{(1)}(\tau) = G^{(1)}(0) \exp(i\omega_0\tau - \tfrac{1}{2}\gamma^2\tau^2). \tag{145}$$

We see that $G^{(1)}$ oscillates with the central frequency of the line and that it decreases for values of $\tau \neq 0$. The envelope of $G^{(1)}$ has a Gaussian sort of behaviour (Fig. 10).

In a photon coincidence experiment we measure $G^{(2)}$. To calculate $G^{(2)}$ we note that $G^{(1)}(x_1, x_1)$ is the function for which τ vanishes so we have $G^{(1)}(0)$

$G^{(1)}(0)$ as the first term of (140),

$$G^{(2)}(x_1x_2, x_2x_1) = G^{(1)}(x_1, x_1)\, G^{(1)}(x_2, x_2) + |G^{(1)}(x_1, x_2)|^2$$
$$= G^{(1)}(0)\, G^{(1)}(0) + |G^{(1)}(0)|^2 \exp(-\gamma^2\tau^2) \qquad (146)$$

The cross correlation function gives the variable contribution. The square of the cross correlation function does not have the oscillating part, it is simply the squared envelope, as it were, of $G^{(1)}$, so we get

$$G^{(2)}(x_1x_2, x_2x_1) = \{G^{(1)}(0)\}^2 \{1 + \exp(-\gamma^2\tau^2)\}. \qquad (147)$$

By plotting $G^{(2)}$ (coincidence rate) against τ (Fig. 11), we see that $G^{(2)} \to 1$ as

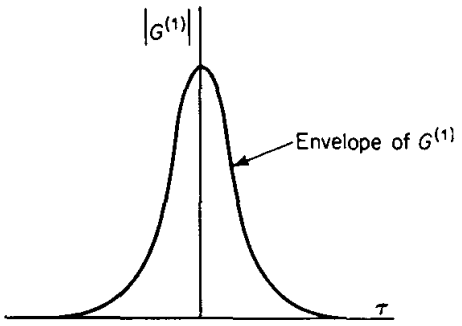


FIG. 10

$\tau \to \pm\infty$ but is 2 at $\tau = 0$. This shows that the HBT effect is not a small effect. In the original experiment it was small because the counters had long resolution times. The HBT effect does not last over a long time interval. The time interval τ is of order of $1/\gamma$ where γ is of the order of the frequency band width, so it helps to have a light beam with very narrow frequency spread to make the effect last for a somewhat longer time. The shape of the curve here is Gaussian because that is the transform of the spectrum we used. Had we used a natural line-shape, then we would have had an exponential behaviour of the HBT correlation on each side of $\tau = 0$ as shown in Fig. 11. Notice that we are

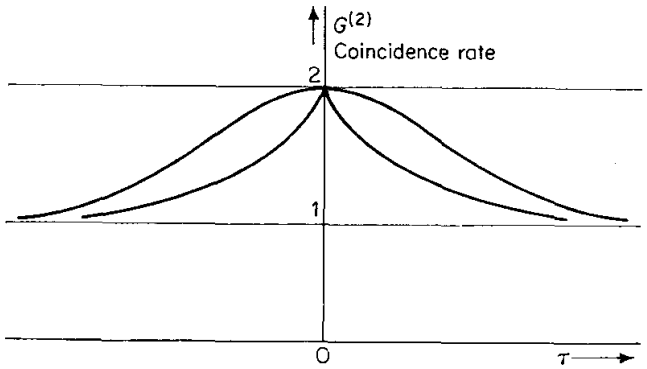


FIG. 11

describing the effect without putting any mirrors in the calculation at all. We are just considering detection of photons in a one dimensional beam with two counters which we assume have infinitely small resolving time. There is always a tendency towards coincidences and this is, in effect, a statement that the photons tend to clump in the beam. We can still ask why this happens. The argument we have given is mathematically complete but it may be that there is not enough in what we have considered to convey the full physical image. In other words, the theorem which says that the higher order correlation functions can be expanded in terms of the lower ones is the essential point of the derivation. Everything we ever need to know about the character of Gaussian stochastic processes really is included in that statement but it is not, perhaps, completely clear what that statement is in physical terms. So let us use a slightly different method. We need not use this fancy expansion to evaluate the higher order correlation function, we can go back to the Gaussian distribution itself and imagine doing the integral. Let us see how that works.

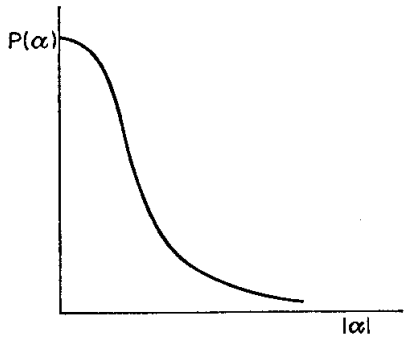


FIG. 12

Consider the correlation functions for $\tau = 0$. The HBT effect is there already; we do not see how the effect relaxes over time, but we do see, for example, that the coincidence rate is twice the square of the ordinary counting rate, eqn. (149). For time interval zero, $G^{(1)}(0)$ is just the second moment of $P(\alpha)$,

$$G^{(1)}(0) = \int P(\alpha)\,|\mathscr{E}|^2 \mathrm{d}^2\alpha = \text{constant} \int \exp\left(-|\alpha|^2/\langle n\rangle\right) |\alpha|^2 \mathrm{d}^2\alpha. \qquad (148)$$

Here for simplicity we have assumed that just one mode of the field is excited. Nothing in the argument really depends on the number of modes that is excited except the relaxation properties. The fact that there are many different modes excited which all have slightly different frequencies obviously determines the way in which the HBT correlation disappears as a function of time, but here we are looking at the effect just at one moment of time, at time interval zero. In that case $G^{(1)}$ is just the second moment of the Gaussian distribution.

The function $G^{(2)}$ is just the fourth moment of the Gaussian distribution,

$$G^{(2)} = \int P(\alpha)\,|\mathscr{E}|^4 \mathrm{d}^2\alpha = 2\left(\int P(\alpha)\,|\mathscr{E}|^2 \mathrm{d}^2\alpha\right)^2 = 2\{G^{(1)}(0)\}^2. \qquad (149)$$

It is a property of Gaussian functions that the fourth moment is twice the square of the second moment. Does that have any physics in it? Well, it is a little hard to call it physics—it is so simple. The Gaussian function has a long tail, even though it decreases quickly (Fig. 12). The fact that the Gaussian has this tail means that the fourth moment is larger than the square of the second moment. If the Gaussian function were to cut off sharply, it would not be possible for the fourth moment to be so very much larger than the square of the second moment. So here we have it. It is because the Gaussian function has a tail that we find that these higher order correlation functions evaluated for time zero are generally larger by a factor of $n!$ than the nth powers of the first order correlation functions. For a coherent field, on the other hand, with random phase, we have essentially a δ-function structure. If we had a precisely determined phase, for a coherent state, then, indeed, we would be considering a δ-function at one point in the complex plane. We essentially never have such information about laser beams; we never know, for example, when precisely they are turned on. So the actual representation of the function P for the most perfect of laser beams would be one in which there is a kind of circular symmetry in the complex α-plane and we have a probability distribution which is zero, except on a cylindrical shell, a shell obtained by rotating the diagram Fig. 13 about its vertical axis. That is what P would look like for the most

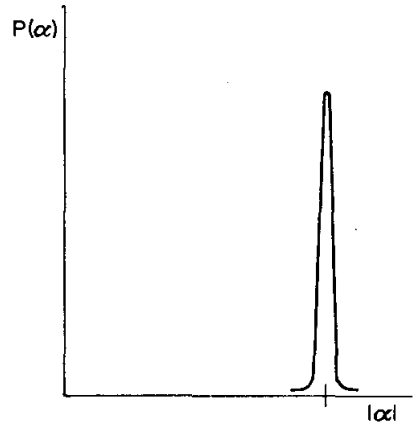


FIG. 13

perfect of coherent states with random phase. Now we see that that kind of distribution has no variance at all in radial terms. For that kind of distribution it is simply true that the fourth power of $\mathscr{E}$ when averaged is precisely equal to the average of $\mathscr{E}$ squared, and squared again; that is why there is no HBT effect for the purely coherent field.

Let us put it in still more elementary terms. In a chaotic field, the amplitude is fluctuating all the time. Sometimes the amplitude is large, more often, the amplitude is small. If we put two photon counters in the field, they will both of them tend to register when the amplitude happens to be large, and they will neither of them tend to register when the amplitude happens to be small. That automatically means that there will be a correlation in the behaviour of the two counters. The two counters do tend to register together and they tend to keep quiet together. No such correlation is present in the coherent field because the amplitude tends to remain constant as a function of time. Both counters are seeing the same amplitude at all times. Not just the same as one another, but an amplitude constant in time. For that reason, their behaviour is statistically independent of one another. Now that leaves us with a number of other questions, of course. It was surmised at an early stage that a laser beam was represented to quite good accuracy by one of these coherent fields. Why should this be so? It is a long story to analyse in detail. One reason is that, to a fair approximation, a laser is a macroscopic distribution of atomic polarizations equivalent to a macroscopic current distribution which does not recoil perceptibly when it emits photons. A laser is, to a good approximation, the kind of thing Kibble described by speaking of the radiation emitted by a specified current distribution (see Kibble, this volume). The situation is one in which the current is specified and the field is quantized, but that is not enough really to characterize the situation physically.

We could still imagine random behaviour in this classical current which would lead to much variance in the amplitude radiated by the field and there is something intrinsic to the laser which tends to suppress that amplitude fluctuation. The laser is a non-linear device which has a kind of feed-back built into it. The field is radiated by a population inversion which has to be maintained in order for the device to keep amplifying its own field. If the field ever gets stronger than a certain steady value, it tends to deplete the inversion, the amplification tends to decrease, and the device tends to stabilize its own amplitude. The theoretical analyses of the laser do not really show that a precisely coherent state is radiated by the device, but they do tend to show that, at sufficiently high power levels, quite a fair approximation to such a coherent state is what is radiated. The complications of dealing with the laser problem itself are quite imposing and are described by Haken in this volume.

We see that to explain the HBT effect quantum mechanically we go back, in essence, to its classical explanation. The important point is that we have found the justification for using that essentially classical explanation in the quantum context.

Those are the best ways of explaining the HBT effect in elementary form. However, every time I present explanations of this sort, I find students come up later and ask, "What about the photons in all of this? What you have done

really is to persuade us that it is not worth talking about photons at all. What you ought to do is go back to the same old classical discussions that were used long ago and simply give them their correct quantum mechanical interpretation." Now, all of us were taught somewhere in the early chapters of the quantum mechanics books that photons are little puffs of probability amplitude; that they have well defined wave-packets that move with the velocity of light and tell us whatever we are capable of knowing about the localization of the photon. In our discussions of photon counting experiments, of the HBT effect in particular, we do not see any of those wave-packets at all. My own feeling is that we are much happier for not having the wave-packets present. Conceptually, the wave-packets are certainly a very good way of looking at the localization of quantum mechanical particles but, in practice, we never know the various parameters that we would have to know in order to determine the wave-packets for individual particles. If a cyclotron produces a beam of charged particles, there is nobody in the cyclotron laboratory who can tell us what the wave-packets are of those emerging particles. What they *can* tell us is the degree of collimation of the beam, the energies of the particles in the beam and general data of that sort. The simplest description of that beam is a description analogous to the kind we have been using here for photons; it is really a density operator description which does not attempt to construct wave-packets for the individual particles because, to do it, we have to deal with information that we simply do not have. On the other hand, at least from a conceptual standpoint, it is an interesting thing to ponder the problem of just how we would go about constructing a description of a beam of particles if we did know everything about the wave-packets of the individual particles. That is what we shall do in the next section for the particular case of a chaotic field.

9. Wave Packet Structure of Chaotic Field[10]

The formula for the chaotic density operator is just the Gaussian mixture of coherent states,

$$\rho = \int \exp\left[-\sum_l (|\alpha_l|^2/\langle n_l\rangle)\right] |\{\alpha_k\}\rangle\langle\{\alpha_k\}| \prod \frac{d^2\alpha_k}{\pi\langle n_k\rangle} \tag{150}$$

Evaluating the Gaussian integrals we obtain,

$$\rho = \sum_{\{m_k\}} \prod_k \frac{\langle n_k\rangle^{m_k}}{(1+\langle n_k\rangle)^{1+m_k}} |\{m_k\}\rangle\langle\{m_k\}|. \tag{151}$$

The density operator is the sum of state vector dyadics corresponding to all

possible numbers of quanta in the individual modes. Because it is a stationary operator and has a separable or factorizable P-representation it is, in fact, diagonal in terms of the occupation number representation. The coefficients of the various quantum states are just powers of elementary parameters. The parameters are just the ratios $\{\langle n_k\rangle/(1+\langle n_k\rangle)\}$ which for the case of thermal radiation, when raised to the power m_k, become

$$\left(\frac{\langle n_k\rangle}{1+\langle n_k\rangle}\right)^{m_k} = \exp\left(-m_k\hbar\omega_k/kT\right). \tag{152}$$

Suppose we are given a collection of as many identical photon packets as we want. How do we put those photon wave-packets together to make the field which is described by the chaotic density operator? When we discover how to do that, we shall learn something about the microscopic structure of chaotic fields. We shall find out, within chaotic fields, something about where the photons are relative to one another and the distribution of their number. A one-photon packet is described by

$$\sum_k f(k)\, a_k^\dagger |\,\mathrm{vac}\rangle. \tag{153}$$

This is the basic element in the calculation. We are going to create a single photon but, we may create a photon in any mode so we sum over all modes, k. It takes a certain set of amplitudes, one amplitude $f(k)$ for each mode in order to specify what the packet is. We can make the packet look like anything we please by appropriate choice of this set of numbers $f(k)$. Imagine that the packet is localized somewhere in space. In fact, it might be localized in a different way. Nothing we are saying really will bear on the question of just how it is localized but imagine that it is a wave-packet that has some particular spatial localization. Let us even assume that that packet is somewhere near the origin $\mathbf{r} = 0$, just for simplicity. This state has to obey a normalization condition, which we obtain by just squaring the state, thus

$$\langle \mathrm{vac}\,|\sum_k f^*(k)\, a_k \sum_{k'} f(k')\, a_{k'}^\dagger |\,\mathrm{vac}\rangle = \sum_k |f(k)|^2 = 1. \tag{154}$$

The square is the sum of the squared moduli of the coefficients $f(k)$. So those coefficients f must obey the normalization (154). Now let us imagine displacing this wave-packet from the origin to the point $\mathbf{r}$. We do that by applying a displacement operator, which in quantum mechanics takes the particular exponential form shown:

$$\begin{aligned}&\exp\left(-i\mathbf{P}\cdot\mathbf{r}/\hbar\right)\sum f(k)\, a_k^\dagger |\,\mathrm{vac}\rangle \\ &\quad = \sum f(k)\exp\left(-i\mathbf{k}\cdot\mathbf{r}\right) a_k^\dagger |\,\mathrm{vac}\rangle \\ &\quad \equiv A^\dagger(\mathbf{r})\,|\,\mathrm{vac}\rangle.\end{aligned} \tag{155}$$

We know what the momentum of any one of these photon states is. Let us assume now that it is a state with a particular propagation vector. Let us assume that the modes k, are plane-wave modes. So to apply the displacement operator means just including in our summation over modes, the particular phase factor associated with that propagation vector. The summation of creation operators which creates a photon packet in the neighbourhood of $\mathbf{r}$ can be written as

$$A^{\dagger}(\mathbf{r}) \equiv \sum_{k} f(k) \exp(-i\mathbf{k} \cdot \mathbf{r}) a_k^{\dagger}. \tag{156}$$

These are the basic elements in the "do it yourself kit". We are given as many as we want of these packet creation operators referring to all possible points in space. The problem is to put these operators together so as to create a chaotic field. The method is not immediately obvious. If we just take a collection of the operators and apply them completely randomly (or in what we think is a completely random manner) to the vacuum state, we shall not, in fact, be creating chaos, we shall be creating order.

The state with n packets identical except that they are shifted relative to one another in position so that they are at the positions $\mathbf{r}_1, \ldots, \mathbf{r}_n$ is given by

$$\prod_{j=1}^{n} A^{\dagger}(\mathbf{r}_j) \mid \text{vac}\rangle. \tag{157}$$

Here we have applied the appropriate succession of creation operators to the vacuum state. It does not make any difference what order we apply those operators in, since the creation operators all commute with one another. In fact, that shows us that we are obeying the necessary symmetry rules; this state is symmetrical under the interchange of the coordinates of all the wave-packets, so we are describing bosons correctly.

We define the normalization function for the state (157) as

$$W^{(n)}(\{\mathbf{r}_j\}) \equiv \langle \text{vac} \mid \prod_{j} A(\mathbf{r}_j) \prod_{l} A^{\dagger}(\mathbf{r}_l) \mid \text{vac}\rangle. \tag{158}$$

Let us describe an essential problem that we encounter in defining the state given by (157). The state is not normalized and the normalization of the state depends on just where the photons are relative to one another. If we are going to normalize the state, we have to calculate the norm of the vector we have written (its square length, in other words). The squared length is written out on the right of (158). It is the vacuum expectation value of a product of packet annihilation operators and packet creation operators. To evaluate the function $W^{(n)}(\{\mathbf{r}_j\})$ it is a good idea to define another function which we can construct

in terms of a single creation operator for a packet at one point and a single annihilation operator for a packet at a different point,

$$v(\mathbf{r}-\mathbf{r}') \equiv \langle \text{vac} \,|A(\mathbf{r})\, A^{\dagger}(\mathbf{r}')\,|\, \text{vac}\rangle. \tag{159}$$

The operator product can be written equally as a commutator because if we consider the two operators in their commuting positions, we then have an annihilation operator on the right which applies to the vacuum state, and it is zero. Since the commutator is a c-number the function v reduces simply to the commutator itself,

$$v(\mathbf{r}-\mathbf{r}') = [A(\mathbf{r}), A^{\dagger}(\mathbf{r}')]. \tag{160}$$

By using the expression (156) and its adjoint we then find

$$v(\mathbf{r}-\mathbf{r}') = \sum_{k} |f(k)|^2 \exp[i\mathbf{k}\,.\,(\mathbf{r}-\mathbf{r}')]. \tag{161}$$

This is the explicit expansion of the function. If we know the packet amplitude $f(k)$ then we know the function v.

The function v, however, does not tell us anything about the phases of the packet amplitudes. It is easy to prove the following properties of function v,

$$\begin{aligned} v(0) &= 1 \\ v(-\mathbf{r}) &= v^*(\mathbf{r}) \\ |v(\mathbf{r})| &\leqslant 1. \end{aligned} \tag{162}$$

Firstly, the normalization condition; if $\mathbf{r} = \mathbf{r}'$, we have just the sum of the squared moduli of $f(k)$, that is, unity. If we invert the argument of the function we get the complex conjugate, obviously. Furthermore, this function is bounded by unity, which is the value it takes on for zero argument, and if we calculate this function, we find, typically, for any smooth behaviour of the amplitudes $f(k)$, the function goes to zero as the points $\mathbf{r}$ and $\mathbf{r}'$ recede from one another. The packet, in other words, would have only a finite size. Any photon packet would have some characteristic packet dimension and v would vanish for $|\mathbf{r}-\mathbf{r}'|$ larger than that particular dimension.

The function $W^{(n)}$ which is the normalization function, is easily seen to be the sum of products given by

$$W^{(n)}(\{\mathbf{r}_j\}) = \sum_{\text{perm}} \prod_{j=1} v(\mathbf{r}_j - \mathbf{r}_{Pj}).$$

We can use the same technique as we used to evaluate high order correlation functions for the Gaussian case to prove this theorem for the normalization functions. What that means then for the first few cases is worth illustrating. For a single photon packet we have

$$W^{(1)} = 1. \tag{163}$$

If we have two packets in the field, $W^{(2)}$ contains the product $v(0)\,v(0)$ that is, just unity, and then we have the additional term with the arguments permuted which just adds $|v(\mathbf{r}_1-\mathbf{r}_2)|^2$; thus

$$W^{(2)} = 1+|v(\mathbf{r}_1-\mathbf{r}_2)|^2. \tag{164}$$

If we have a state with three packets in it we have a somewhat more complicated structure,

$$W^{(3)} = 1+|v(\mathbf{r}_1-\mathbf{r}_2)|^2+|v(\mathbf{r}_2-\mathbf{r}_3)|^2+|v(\mathbf{r}_3-\mathbf{r}_1)|^2 + 2\,\mathrm{Re}\,v(\mathbf{r}_1-\mathbf{r}_2)\,v(\mathbf{r}_2-\mathbf{r}_3)\,v(\mathbf{r}_3-\mathbf{r}_1). \tag{165}$$

These two cases $W^{(2)}$ and $W^{(3)}$, illustrate the structure of the normalization function $W^{(n)}$ in its simplest form. If the packets are far apart, for example, $\mathbf{r}_1$, is very different from $\mathbf{r}_2$, then the overlap amplitude v vanishes and if all of the packets are far apart in the n-fold case and tend not to overlap, then all of these normalization functions are going to reduce, essentially, to unity $W^{(n)} \approx 1$. The normalization functions, however, tend to differ from unity when the packets are made to overlap. When $|\mathbf{r}_1-\mathbf{r}_2|$ is small enough so that the packets do overlap, for example, the function v rises to its maximum value of one, and, more generally, we find that the maximum value of $W^{(n)}$ is $n!$. So that

$$n! \geqslant W^{(n)} \geqslant 0. \tag{166}$$

If we imagine distributing wave packets at random positions in the field, we shall find the normalization function is essentially always one, or nearly always one if we have a very dilute photon gas. But as the packets begin to overlap, the normalization moves away from unity and every time two packets precisely overlap there is a bump in the normalization function. We have to take that into account of course if we want to deal with a correctly normalized state. One of the hazards of dealing with wave-packets is that calculations based on them have not always been done with correctly normalized states.

A correctly normalized state for wave-packets is given by

$$|n\{\mathbf{r}_j\}\rangle \equiv \frac{1}{\sqrt{W^{(n)}(\{\mathbf{r}_j\})}} \prod_j A^\dagger(\mathbf{r}_j)\,|\,\mathrm{vac}\rangle. \tag{167}$$

We have taken the state generated by the n packet creation operators and have put in the reciprocal of the square root of our normalization function.

In our process of constructing the chaotic field we naturally want to put these photon packets in all possible different positions in the field, that is, we naturally think of a chaotic field having properties we assume to be uniform as a function of spatial variables. We wish to consider a mixture of states in which these photon packets are in all possible positions. A pure state with one

particular set of positions for the photon packets is given by (167). We mix these states together by multiplying by some kind of a weight function of the photon packet coordinates, then integrate over all photon packet coordinates. We know that the total number of photons in a chaotic field is quite indefinite because, in general, we do not know how many photons there are in any one mode so we have only a statistical notion of how many there are in the entire field. We therefore have to use a succession of these weight functions, one for every value of n and sum over all possible values of the number of photons in the field. The density operator we construct is given by

$$\rho = \sum_n \int F_n(\mathbf{r}_1 \ldots \mathbf{r}_n) \, |n\{\mathbf{r}_j\}\rangle\langle n\{\mathbf{r}_j\}| \frac{\Pi d\mathbf{r}_j}{V^n} . \tag{168}$$

We now wish to know the kind of weight functions we should choose in order to obtain a chaotic field. In other words, we wish to generate real chaos, to scrupulously avoid order, to avoid having the entropy of the field at anything less than its maximum value. The crucial point is the location of the photons to maximize entropy. At this point it should be noted that we can prove that the correct weight function for a chaotic field is

$$F_n(\mathbf{r}_1 \ldots \mathbf{r}_n) = c_n W^{(n)}(\mathbf{r}_1 \ldots \mathbf{r}_n), \tag{169}$$

where c_n is a constant. The proof is a bit long and is therefore omitted, but basically one way of proving it is as follows. We take the matrix elements of ρ between coherent states, and go back to the original chaotic Gaussian density operator (150) and take the matrix element of that between coherent states and then compare the two expressions obtained. We want them to assume the same form and the only way they can ever have the same form is by taking these weight functions F_n to be proportional (the constant of proportionality is c_n which is independent of packet positions) to the normalization function $W^{(n)}$. Now that is simply a mathematical result and the only way of justifying it is by going back and making the explicit comparison and showing algebraically that that is the only way we can do it. But we have there an interesting statement about the microscopic structure of a chaotic field because this function F_n is a weight function telling about the tendency of photon packets to overlap, (whether they really tend to overlap or not). The statement is here that they must have a distinct tendency to overlap. The function $W^{(n)}$ is something which is unity when no photon packets overlap. However, every time a pair of photon packets overlaps it jumps away from the value unity to a larger value. In other words, this weighting tends to favour the overlap of wave packets. Many things are said about the HBT effect. A number of authors state that it is purely and simply a consequence of Bose–Einstein

statistics. We can see here that that statement is an extremely incomplete one, and if that is the only statement which is made, it is incorrect. It is incorrect because we can imagine photon wave-packets having any distribution of positions we please. Boson statistics are extremely permissive, we can imagine constructing states with any kind of anti-correlation we like between photon packets. How we would generate those states and what the practical device is that would do it, is another question. It might be very difficult to think of a device which would create anti-correlation between photons.

The essential point is this. For Boson statistics and a requirement of maximum entropy, then the only way of meeting the requirement is to give the photon packets a distinct tendency to overlap and use the weight function $W^{(n)}$ in this construction which gives us an additional unit of weight every time two packets overlap. If we use this weight function in doing the calculation, then the density operator obtained is

$$\rho = \sum_n c_n \int A^\dagger(\mathbf{r}_1) \dots A^\dagger(\mathbf{r}_n) |\text{vac}\rangle\langle \text{vac}| A(\mathbf{r}_n) \dots A(\mathbf{r}_1) \frac{\Pi \, d\mathbf{r}_j}{V^n}. \tag{170}$$

The function $W^{(n)}$ put in as a weight, cancels the reciprocal of $W^{(n)}$ which is inserted in order to normalize, and so the density operator has a very much simpler structure. It has precisely the sort of structure that we would have guessed if we had guessed wrongly and forgotten all about the normalization problem to begin with. That is what has happened in some of the papers that have been written on the subject which purport to derive the HBT effect. It is one of the reasons why they say it results only from Boson statistics. What has happened is that, without any attempt at justification, they have used a structure like (170) which is not normalized (it has a variable normalization). In fact, the HBT effect has been put into the very first statement of the derivation, and, naturally enough, it is taken out at the end.

We still have to determine the coefficients c_n, and it is not clear at this stage what the relationship is between our packet amplitudes $f(k)$ and the mean occupation numbers n_k. It is easy enough to derive these relationships. We simply carry out the spatial integrations on the form of the density operator given by (170) and we obtain the following expression in terms of the n quantum states,

$$\rho = \sum_{n=0}^{\infty} n! \, c_n \sum_{\Sigma m_k = n} \prod_k |f(k)|^{2m_k} |\{m_k\}\rangle\langle\{m_k\}| . \tag{171}$$

This is a double summation, a summation over the total number of quanta in the field and a summation over the set of occupation numbers of the individual modes. In the first summation the total number of quanta in the field has to

be restricted to the value n which is then summed over its full range from zero to infinity. We now compare this expression with the one that goes back to the chaotic density operator (150). In (171) we see powers of the squared modulus of the packet amplitude. In the original one we had powers of the parameter $\langle n_k\rangle/(1+\langle n_k\rangle)$ where $\langle n_k\rangle$ is the mean occupation number. Obviously these two have to be proportional to one another. So the identification of the one density operator with the other requires that

$$\langle n_k\rangle/(1+\langle n_k\rangle) = Z|f(k)|^2 \tag{172}$$

where Z is the constant of proportionality. Then to complete the identification, the comparison of (171) with (150), we have to say that the unknown coefficients c_n which occur in the weight functions are given by

$$\begin{aligned} c_n &= \frac{Z^n}{n!}\prod_k \frac{1}{1+\langle n_k\rangle} \\ &= \frac{Z^n}{n!}\prod_k (1-Z|f(k)|^2). \end{aligned} \tag{173}$$

The parameter Z is then given by the normalization condition

$$\sum_k |f(k)|^2 = 1 \Rightarrow Z = \sum_k \frac{\langle n_k\rangle}{1+\langle n_k\rangle}.$$

This means that the mean occupation number and the squared packet amplitude have slightly different shapes as functions of frequency or wave number as shown in Fig. 14. When we put the first photon packet in the field we may think that the spectrum of the field eventually is going to look like $|f(k)|^2$, but it will not because of the effect of wave-packet overlap. There will

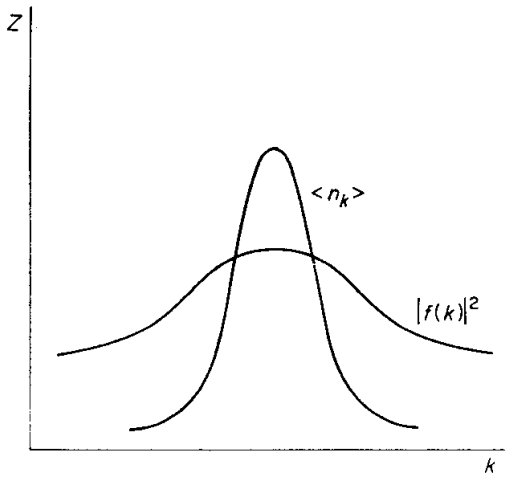


FIG. 14

be the relationship we have written between $|f(k)|^2$ and the mean occupation number, which is a non-linear relationship.

Finally, when we put all of this together, we have the following two expressions for the density operator. One in terms of the wave-packet amplitude squared and the n quantum states and the other in which we see explicitly the sum of the averages of the pure states, which correspond to the presence of packets at all possible positions,

$$\rho = \prod_k (1 - Z|f(k)|^2) \sum_n \frac{Z^n}{n!} \sum_{\Sigma m_k = n} \prod_k |f(k)|^{2m_k} |\{m_k\}\rangle\langle\{m_k\}|$$

$$\rho = \prod_k (1 - Z|f(k)|^2) \sum_n \frac{Z^n}{n! V^n} \int \prod_j A^\dagger(\mathbf{r}_j) |\text{vac}\rangle\langle \text{vac}| \prod_j A(\mathbf{r}_j) \prod_j d\mathbf{r}_j. \quad (174)$$

Now can the latter expression be used for anything?

Its principal use has been given above and lies in the statement that, in constructing a chaotic field from wave-packets, it is necessary to put the packets together with a weighting $W^{(n)}$ which tends to favour the overlap of packets. We can use this form of the density operator for various sorts of calculations, for example, it is a relatively convenient form for finding the distribution of the number of photons in the field. But if we try doing photon counting calculations with this representation of the density operator, we find that the complications are immense. For example, to describe the absorption of a single photon from the field we have the problem that the photon may be absorbed from any of these packets and just to describe the single, most elementary process of absorption of one photon, we would have to take into account interference terms in all of these packets because we generally do not know from which packet the photon was absorbed. So the use of photon packets does lead to immense algebraic complications in actual calculation and it is something to be avoided. On the other hand, we *can* carry through the comparison of the density operator formalism that we have used with the photon packet scheme and in that way see something about the microscopic structure of the field.

One of the advantages of this approach is that the treatment of Fermions is really no different in principle from the treatment of Bosons. That is one of the real gains in using this particular approach, because we cannot use the coherent state approach at all in discussing Fermion beams. There is simply no such thing as a coherent state for Fermions. For any Fermion mode we can only have the occupation number zero or one, and there is no such thing as an eigenstate of the annihilation operator. An eigenstate cannot be constructed with those two possibilities. Try doing it algebraically and you will see what I mean. There are no coherent states for Fermions but it is easy to construct

maximum entropy states for Fermions. We can do that just by inspection. Since we are specifying the mean number of Fermions present, we are saying, in other words, that $\mathrm{Tr}(\rho a^\dagger a)$ for one mode is equal to a certain fixed expectation value,

$$\mathrm{Tr}(\rho a^\dagger a) = \langle n_k \rangle, \qquad n_k = \begin{cases} 0 \\ 1 \end{cases} \tag{175}$$

That means, as part of the maximization process for entropy, that the density operator has to be diagonal, so the density operator must be a linear combination of the projection on the zero quantum state for one mode and the one quantum state. It must have the specified expectation value, and it must have trace unity. We have only two unknown coefficients and that determines the two coefficients, so in fact, the density operator has to be this:

$$\rho = (1-\langle n \rangle)\,|0\rangle\langle 0| + \langle n \rangle |1\rangle\langle 1| \quad \text{where} \quad \mathrm{Tr}\,\rho = 1. \tag{176}$$

This may be generalized to fields containing many modes by the same arguments as for Bosons and we find the following relation between the occupation numbers for Fermions and the packet amplitudes,

$$\frac{\langle n_k \rangle}{1-\langle n_k \rangle} = Z\,|f(k)|^2 \Rightarrow Z = \sum_k \frac{\langle n_k \rangle}{1-\langle n_k \rangle}, \tag{177}$$

where the coefficient Z is determined again from the normalization condition. The density operator obtained, finally, has almost the same structure as that for Bosons,

$$\rho = \prod_k \frac{1}{1+Z|f(k)|^2} \times \sum_n \frac{Z^n}{n!V^n} \int \prod_j A^\dagger(\mathbf{r}_j)\,|\,\mathrm{vac}\,\rangle\langle\,\mathrm{vac}\,|\prod_j A(\mathbf{r}_j) \prod_j \mathrm{d}\mathbf{r}_j. \tag{178}$$

As far as the appearance of the summation over states in which we have used the packet amplitudes is concerned, that is precisely the same. The normalization factor simply looks a little different. The point is that the operators $A(\mathbf{r})$ anti-commute, and so the function v is the anti-commutator of the two operators, the one for creation and the other for annihilation of a packet,

$$\{A(\mathbf{r}), A^\dagger(\mathbf{r}')\}_+ = v(\mathbf{r}-\mathbf{r}'). \tag{179}$$

The normalization function $W^{(n)}$ is an antisymmetrical sum over permutations of products of the functions v,

$$W^{(n)} = \sum_P (-1)^P \Pi v(\mathbf{r}_j - \mathbf{r}_{Pj}). \tag{180}$$

That means that the packets will tend to avoid one another. This is a normalization which will go to zero whenever two packets overlap. In other words, in attempting to put together the chaotic density operator for Fermions with our "do it yourself kit" we lose every time we set down two particle packets overlapping each other. We are only allowed to put states together with packets not overlapping and, indeed, there is an anti-HBT effect for Fermions, and that *does* come purely and simply from the Fermi statistics. That would be true whether we were considering a chaotic state for the Fermions or any other state. What we have shown is that a chaotic state for Fermions does have a particularly simple representation which is completely analogous to the one which we derived for Bosons.

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Photon Statistics †

E. R. PIKE

INTRODUCTION

In this chapter I shall be discussing the statistical properties of optica radiation fields. I imply immediately that we are interested in describing light fields in terms of randomly fluctuating quantities, and that statistical correlations between these random quantities are experimentally measurable and amenable to theoretical interpretation. I should like first of all to spend some time explaining why, since optics is such a venerable subject in physics, such statistical considerations seem to be comparatively recent. It is, nowadays, well realised that if we consider, for instance, one component of the electric field vector produced at a point in space by a classical (thermal) light source, this is a violently fluctuating quantity. Thinking in rather loose terms for a moment, we may consider the intensity of such a source at this point in space. This is a random variable whose most probable value is, in fact, zero. Thus, if I were to place my eye behind a sufficiently small aperture at this point in space, and look at the source, then most of the time I should see darkness. The reason why physicists have been able to avoid considering the true nature of such sources and to construct the whole of classical optics theory as though lamps were of constant brightness, lies in the time scale of these intensity fluctuations. The fluctuations are slower, the more monochromatic the source, but even for the most monochromatic classical sources available, the time over which these fluctuations take place is still very small —in the region of a few nano-seconds at most. The origin of the fluctuations is simply the random beating together of wave trains from various parts of the source, which are spontaneously emitted and uncorrelated in phase. At the present time, one can construct a source with the same statistical properties as a classical lamp merely by scattering laser light from a medium whose dielectric constant is undergoing random but slow fluctuations, and one can

† The material presented here is a slightly expanded version of a review by the author published in "The Growth Points of Physics," *Rivista del Nuovo Cimento,* Vol. 1, Numero Speciale 1969, and is reproduced by kind permission of the publisher.

indeed see with the eye the properties which I have just described. A simple example is laser scattering from micron-size polystyrene balls suspended in water and undergoing Brownian motion. The time scale of the intensity fluctuations in this case is of the order of one-tenth of a second.

In spite of the short time scale of the intensity fluctuations from thermal light sources, the first successful statistical experiments were performed using such sources, some years before the laser was invented. These were the experiments of Forrester, Gudmundsen and Johnson,[1] and of Hanbury-Brown and Twiss.[2] These were difficult and pioneering experiments at the time, but the development of the photomultiplier tube as a detector, and fast transistor-electronic circuitry for data processing, makes such experiments very much easier today. These early measurements of intensity fluctuations of optical light sources are so often discussed and quoted in relation to this subject that, rather than dealing with them in the appropriate place in the general scheme of this chapter, it is worthwhile in this introduction to say essentially what was measured, and to try to make quite clear exactly what the Hanbury-Brown–Twiss effect really is. First of all let me make plain one thing that the Hanbury-Brown–Twiss effect is not; it is not a tendency of photons to bunch in time, solely because they are bosons. Much of the discussion following the Hanbury-Brown–Twiss experiments would have been clearer had the laser existed at the time. A laser is a source of photons which show no bunching properties whatsoever. If a photodetector is placed in the field of an ideal laser, photodetections will take place completely at random, with no correlation between adjacent events. The existence of such a source shows that the intensity correlations observed in the experiments of Hanbury-Brown and Twiss and of Forrester *et al.* are not directly attributable to the boson nature of photons.

In fact a completely classical version of these experiments was developed during the Second World War in the field of radar meteorology. The statistical properties of the electric field of a scattered radar beam are the same as those of the electric field produced by a classical lamp. We can imagine that the scattering of a radar beam by a cloud of randomly positioned rain drops moving in the wind, might be similar to the scattering of a laser beam by a random set of Brownian particles, or more exactly to laser scattering from particles suspended in a flowing fluid. The common feature is a Gaussian distribution of (complex) field amplitudes. The extraction of spectral information from the intensity fluctuations of the scattered radar beam presents exactly the same problem as the determination of the properties of the particle motion in the laser-scattering experiment, or the explanation of the Hanbury-Brown–Twiss effect. The same formula—eqn (49) below—applies in all these cases.

The explanation of the radar experiment is quite simple. Consider the

intensity received from a single raindrop illuminated by a constant intensity beam—it will also be constant. Now consider what happens if two raindrops moving at a fixed relative velocity are illuminated simultaneously. The received beams will be at different Doppler-shifted frequencies and the intensity received will fluctuate due to the beating together of the two return signals. The time scale of the intensity fluctuations gives information about the relative motion of the particles. If we now consider a whole set of particles in random relative motion, the same general ideas apply. Each particle produces a Doppler-shifted return signal which beats with all the others. The spectrum of intensity fluctuations is then the fold of the spectrum of Doppler shifts with itself, and the normalized autocorrelation function of the (stationary) intensity fluctuations $g^{(2)}(\tau) = \langle I(\tau)I(0)\rangle/\langle I(0)\rangle^2$ is just the Fourier transform of this, and has the general form shown in Fig. 1.

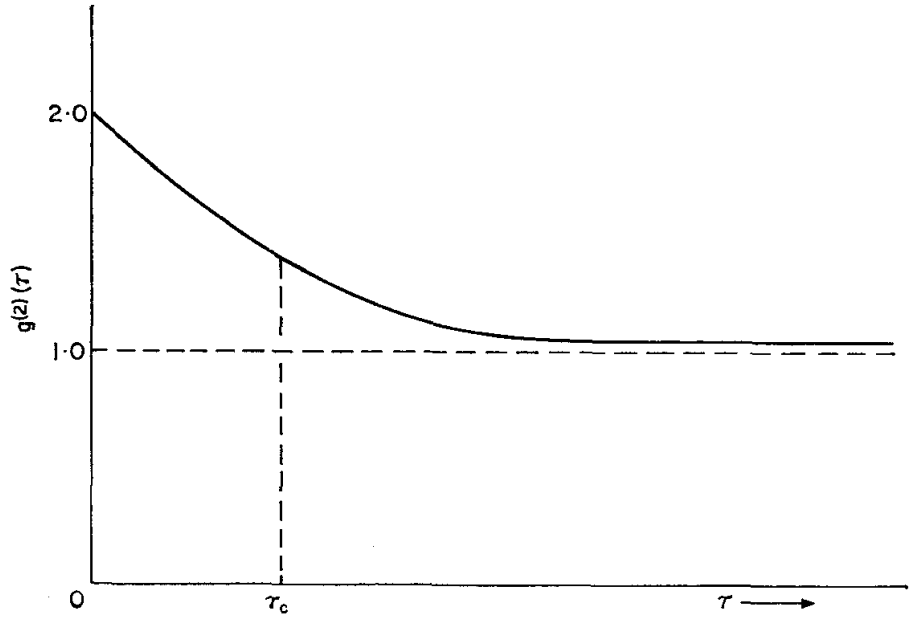


FIG. 1. General form of the normalized autocorrelation function of intensity fluctuations of either radar returned from a random target, of scattered laser light from a fluctuating dielectric medium, or of light emitted by a thermal light source.

The same form is obtained (with essentially the same explanation) for the intensity correlations of a chaotic light source, as measured by Forrester *et al.* and by Hanbury-Brown and Twiss. The source presents to the detector a randomly varying (Gaussian) electric field, with a "coherence time" (suitably defined with respect to the spectral profile) of the order of the inverse of the spread of frequencies present. It should be said, however, that this classical explanation of intensity correlations does not mean that the spectral profile of a source necessarily has a classical origin. The same type of inten-

sity fluctuations would arise from a source which exhibited only the "natural" line width associated with spontaneous emission, which requires a quantum mechanical derivation. Only a very fine distinction can be drawn between the "classical-like" intensity fluctuations of an inhomogeneously (say Doppler-) broadened source, and the "quantum-mechanical" intensity fluctuations arising from the natural line width of an atomic transition, in thermal equilibrium with its radiation field.

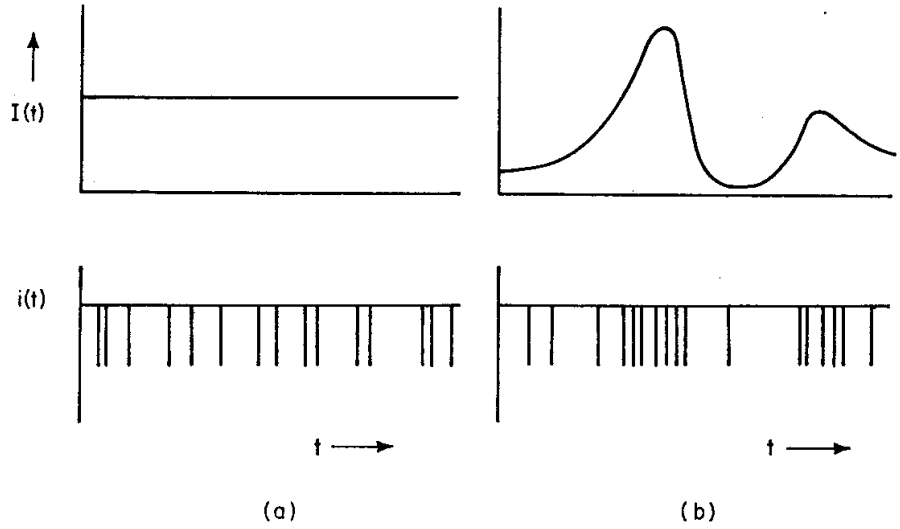


FIG. 2. Illustrating the "bunching" of photodetections due to a fluctuating light intensity. $i(t)$ is the detector output current.
(a) constant intensity, random arrivals,
(b) fluctuating intensity, bunched arrivals.

The association between the correlated intensity fluctuations of a chaotic classical optical source and the "bunching" in time of events recorded by a photodetector placed in the field, is easily explained with reference to Fig. 2. The probability of a photodetection is proportional to the "intensity" of the field (a more comprehensive treatment of photodetection will be given later). For a constant-intensity field such as a perfectly coherent, single-mode laser, the photodetections occur at random in time, while for a fluctuating field the obvious departure from randomness can be called "bunching". The Hanbury-Brown–Twiss bunching effect is thus a quite simple phenomenon. The effect of the usual beam splitter, by the way, is not at all fundamental. It merely serves to expose two detectors to the same light field at zero space and time difference. For measuring correlations between intensity fluctuations at different times at the same space point, one may just as well use a single detector. It gives identical results.

The subject has developed greatly since the days of these early experiments, and statistical considerations now play a central role in many experiments

which use intensity fluctuations, for example, in spectroscopy and high resolution interferometry.

Theoretical consideration of the statistical properties of the general optical field has in recent years led to an improved framework for the theory of optical-field correlations or "optical coherence". The particular case of laser fields has, of course, received an enormous amount of attention, and my final introductory remark concerns the development of the theory of the laser. The laser appeared at an opportune time in theoretical physics, when one of the most interesting and active areas has been the development of techniques for dealing with irreversible processes. A notable date here was about fifteen years ago when van Hove[3] published his work on the derivation of quantum-mechanical master equations. Since these early papers, a great deal of progress has been made, and a vast literature now exists on the general subject of quantum mechanical systems interacting with thermal reservoirs, or quantum statistics. We can also include here the progress in finite-temperature many-body theory. When the quantum mechanical system is a radiation field interacting with non-relativistic states of bound electrons, the subject is variously called quantum optics, quantum electronics, statistical optics or photon statistics. It perhaps could be differentiated from the established discipline of quantum electrodynamics (even in its non-covariant low-energy form) by the emphasis placed on mixed (statistical) states of coupled electron–Maxwell fields and on the influence of dissipative processes, rather than on the pure states of the coupled fields. The progress in this theoretical area has thus been directly applicable to the laser problem, where the treatment of dissipation has been crucial to success.

To review the subject, I shall first spend some time showing how one describes an optical field from the theoretical point of view, placing particular emphasis on the statistical description. Following this, I shall discuss how one describes such a field from the experimental or detection point of view. After these general considerations, detailed results will be given for particular types of field, namely, thermal fields, laser fields and mixed fields. In the final section I shall discuss the practical applications; these make up the relatively new subject of statistical spectroscopy.

1. The Description of a Light Field

In some ways the theoretical techniques required for this subject are generalisations of quantum electrodynamics, since in this field the statistical operator or mixed states are rarely mentioned. The generalisation, however, is not fundamental, and to a far greater extent the theoretical apparatus required is a very restricted subgroup of the known results for the coupled electron–Maxwell fields.

I shall not try here to cover exhaustively all the theoretical papers which have been written on the subject, but try instead to extract from them the essential details of theories which are directly applicable to experiment. This is a much easier task, but only so because of the efforts of many individual workers. I shall mention here particularly the work of Professor Glauber of Harvard University and his students, Professor Haken of Stuttgart and his students and co-workers, Professors Lamb and Scully of Yale University and M.I.T., and Dr. Lax of Bell Telephone Laboratories and his co-workers. It will be impossible to mention individually all contributors to the subject and I shall append general references covering the work of the above-mentioned authors.[4, 5, 6, 7] An excellent pre-1965 bibliography is given by Mandel and Wolf.[8]

It will perhaps help at this stage to look more closely at a typical experiment before embarking on the necessary theory to describe it. In any experiment in optics there exists a source which emits the radiation, the radiation itself which carries energy from the source through some medium which may absorb or scatter (including any optical instrument, for dispersion or diffraction), and finally one or more detectors which absorb the energy of the radiation field in some way which renders it visible. In order to discuss such optical experiments on an intelligent and fundamental level we, as always in physics, consider simplified situations where the (theoretically) inessential complications of the real world are removed as far as is possible. For a detector then, for instance, we postulate an ideal device which renders visible, without noise or distortion, the absorption of energy from the optical field. There are two features of the real world which this ideal detector must incorporate, in order to be a useful approximation to a real detector. First of all, because of the relatively high energy of the optical photon, the output of the detector is not continuous. A real detector does not absorb power from the field continuously, but only in units of one quantum of energy. The second fact is that all the incident power cannot be detected with 100% efficiency. We therefore assume that our detector has a random chance η of detecting each quantum. Consider an experiment in which we set up a source, apparatus and a single detector. The output of the experiment, which we have to describe theoretically, is then a train of single events in time corresponding to the instants at which quanta of energy were absorbed from the field. We do not usually think of the output of an optical experiment in this way, for two reasons. Firstly, in many experiments so much energy is taken from the field that the energy of a single photon is small by comparison, and secondly the area presented to the field by the detector is in most cases so large, that it should really be considered to be a large number of independent detectors in parallel, each of which is absorbing a train of energy quanta. Basically, however, the raw data of optical experiments is in the form of a sequence of

events, and to discuss such experiments accurately in other terms, for example, in terms of a classical continuous detected light "intensity", is possible only in the types of limiting situations mentioned above. From our typical experiment, therefore, we have, perhaps on an oscilloscope screen, a train of pulses in time. How are we to interpret them to compare with theory? Each individual pulse has clearly little significance and we thus need to look at collective properties such as, the mean rate over a certain interval of time (the closest one can get to a classical definition of intensity), any bunching properties, any periodicity, etc., in other words only the statistical properties of the sequence of events are useful.

The fullest theoretical description, therefore, of optical experiments must be fundamentally statistical in nature, predicting such properties of distributions of photodetections. Less complete theories have heretofore been sufficient, but for the reasons outlined in the introduction, interest is now focussed on these more fundamental questions.

1.1 BASIS STATES AND INTERACTION WITH MATTER

With this motivation, we proceed to an elementary theoretical introduction. It will suffice, in exposing the basic results of the subject, to consider in detail the interaction between the free radiation field and a single bound electron only. The free radiation field without either sources or detectors is of trivial interest. The bound electron, for most purposes, may be considered to have only two active energy levels. The effects of many such electrons will be simply additive. The radiation field itself may also, for the purposes of clarifying basic issues, be considered to consist of energy in a single mode, that is to say, we concern ourselves solely with the occupation of a single cell of the radiation phase space, and we may proceed a long way by taking the special case where this is resonant with the electronic frequency. We treat the interaction non-relativistically, in the electric-dipole approximation. With these simplifications we may clearly neglect any consideration of longitudinal and scalar photons, and since no essential detail is lost we may also confine the theory to one state of polarization; we can take the polarization direction, for simplicity, in the direction of the electronic dipole. Thus we are left with the radiation field of one transverse photon. The interaction between this field and the electron is sufficiently weak for us to use the stationary states of the uncoupled fields as a convenient basis set for the description of the combined system. The coupling will then give rise to continuous transitions between these states. At any instant, therefore, the occupation of these basis states describes the state of the interacting system.

Let us denote the two possible states of the bound electron by the ket vectors $|b\rangle$, where b can be b_u or b_l designating the upper and lower levels,

respectively. The radiation field will be specified by the infinite set of states $|n\rangle$, denoting the number of quanta of energy present in the mode volume. The direct product of these two spaces gives the basis set $|r\rangle = |n, b\rangle$ for the combined system. The second quantized operators $b^\dagger$ and b and $a^\dagger$ and a, will create and annihilate the electron and single-photon states respectively.

In this representation, the complete quantum-statistical specification of the state of the system is given by the density operator

$$P = \sum_{rs} P_{rs} |r\rangle\langle s| \tag{1}$$

or, equivalently, by the set of density-matrix elements

$$P_{rs} = \langle r|P|s\rangle. \tag{2}$$

If P_{rs} factorises ($P_{rs} = P_r P_s^*$), then we have a pure state; otherwise, we have a statistically weighted mixture. The general problem requiring solution is, given P_{rs} at time zero, find P_{rs} at time t. Generalized master equations will provide the tools for this task. Statistically stationary states will be found in which P does not change with time, implying that the total transition rates into and out of any given basis state exactly balance.

The interaction Hamiltonian is simply, in second quantized form,

$$V = g[b_u^\dagger b_l a + b_l^\dagger b_u a^\dagger], \tag{3}$$

where the coupling constant g is given by

$$\mu\left(\frac{1}{2}\frac{\hbar\omega}{\Omega}\right)^{\frac{1}{2}}, \tag{4}$$

μ is the electronic dipole moment and Ω the mode volume; antiresonant terms have been neglected. To obtain the state of the radiation field alone, we reduce the total density operator P over the electron states, by simply summing over b_u and b_l, to obtain a reduced density operator ρ. Likewise a reduced density operator σ may be found for the electron, by summing over the states of the radiation field.

To include dissipation in general forms of the theory, the electron system is coupled to a heat bath, which may be at a positive temperature (damping) or at a negative temperature (pumping). Similarly the photon field is coupled to a reservoir which causes decay. However, photon statistics, sufficiently general for this presentation, will result if we confine the external influences to a mechanism for maintaining, on a time scale short compared with variations in the radiation field, fixed probabilities σ_{uu} and σ_{ll} for the occupation of the upper and lower electron states respectively, and a "window" in the volume Ω, which lets out photons at a constant rate, thus damping the radiation field. The former mechanism is required to describe thermal

radiation statistics. The addition of the latter mechanism, as the temperature of the electron system is raised through infinity to negative values, changes the photon statistics to those characterising laser fields.

I have mentioned that the above representation is a convenient one. It is not of course the only possible one and although I shall use it myself in this chapter, I must mention here that the most widely used alternative representation uses an over-complete set of the "coherent" eigen-states of the annihilation operator of the radiation field,

$$|\alpha\rangle = \exp(\alpha a^\dagger - \alpha^\dagger a)|0\rangle$$

$$= \exp(-\tfrac{1}{2}|\alpha|^2) \sum_n \frac{\alpha^n}{(n!)^{\frac{1}{2}}} |n\rangle. \tag{5}$$

So that

$$a|\alpha\rangle = \alpha|\alpha\rangle. \tag{6}$$

As shown by Glauber (see his chapter in this volume), unique expansions in this representation are possible, although, as might be expected, manipulations must be carried out with some care; the treatment of quantum noise, for instance, although possible, and favoured by a number of eminent authors, seems much more difficult than in the number representation. This is only to be expected, in fact, since, as I will mention below, the representation is more useful for the classical limit.

The feature of this representation which is most important, is the convenience with which operators in normal order may be evaluated. The representation also makes the classical limit very easy to obtain; this becomes apparent when van Hove- or Pauli-type master equations are transformed into the α representation, since they then take the form of Fokker–Planck equations for an apparently classical distribution function $P(\alpha)$. The function is not, of course, a classical one in general, and in some cases does not exist in any sensible class of functions. I have not time to go into these interesting questions of formalism but refer you to recent work of Glauber and Cahill,[9] in which the whole subject of quasi probability distributions and the relation between the classical and quantum domains is very expertly treated.

1.2 TIME DEVELOPMENT

To return then to the general problem posed above, namely, given the initial state of the light field and the electron, how does the system evolve?

The required time development can be found in many ways. Traditionally light absorption, for instance, is treated by degenerate secular perturbation theory and Fermi's golden rule. Kelley and Kleiner[10] use the Liouville–Neumann expansion of the time-development operator. Mollow[11] uses

Hamiltonian equations of motion for the individual second-quantized field operators. The Langevin equations or Fokker–Planck equations of some laser theories could even be applied for this purpose. In this chapter I should like to bring to your notice the recent progress which has been made in perturbation theory, in connection with the density matrix itself. The von Neumann equation of motion of the density matrix might be called the very basis of quantum-statistical mechanics, and very early in the history of the subject, Pauli[12] conjectured that equations connecting diagonal elements only and having a gain–loss structure typical of a Boltzmann equation should exist in certain circumstances.

Thus one would initially know the occupation probability of a given level, and as time proceeded, this would be depleted by transitions into the other levels and augmented by similar reverse transitions, at known rates dependent on the perturbation. Such an equation is known as a master equation and, if it can be found, it clearly simplifies the whole structure of quantum-statistical perturbation theory, for any given problem. Indeed in many areas of physics, the Boltzmann equation is used continually for the most detailed practical calculations. The question of the possibility of constructing such an equation, by a fundamental derivation using the von Neumann equation, has been the subject of many papers in recent times. Apart from van Hove, whom I have already mentioned, notable work has also been published by Prigogine,[13] Swenson[14] and Zwanzig.[15] The present situation is very clear but perhaps not yet sufficiently generally appreciated, due to the very technical methods used for most of this work. I can summarize it by stating that equations of such a form can now be written down for all elements of the density matrix which, under the sole condition that the initial density matrix is diagonal, are as general as the von Neumann equation itself. They amount, in fact, to no more than a reformulation of the machinery of quantum mechanics. Thus for instance, for diagonal elements, we have the prescription for degenerate states (Pike[16])†

$$P_{rr}(t) = P_{rr}(0) + \sum_s \frac{1}{i} \int_0^t M_{sr}(t-t')[P_{ss}(t') - P_{rr}(t')]\, \mathrm{d}t', \tag{7}$$

where

$$M_{sr}(t) = -2(-it)V_{rs}V_{sr} + \frac{(-it)^2}{2!} \sum_q (-3V_{rq}V_{qs}V_{sr} + 3V_{rs}V_{sq}V_{qr}) + \text{etc.} \tag{8}$$

The off-diagonal terms also have simple expansions of similar form, but now

† Equation (56) of Pike[16] applies inadvertently to degenerate perturbations only; eqn (60), however, is completely general.

include energy-level differences. It is not difficult, using these formulae with a small density of states spread $h(E)$, introduced in the usual way on to either the radiation mode or the electronic levels, to perform the integrals over energy and time, and obtain the result

$$P_{rr}(t) = P_{rr}(0) + \pi h_0 t \sum_s V_{rs} V_{sr} [P_{ss}(0) - P_{rr}(0)] + O(V^3). \tag{9}$$

This will hold for a time short compared with the transfer rates; h_0 denotes the value of $h(E)$ at the resonant energy.

I refer to my own paper quoted above,[16] for details of these master equations, but can give here a simple method for deriving them.

By definition

$$H = \sum_q E_q |q\rangle\langle q| + \sum_{pq} V_{pq} |p\rangle\langle q|$$

$$P_{rs}(t) = \mathrm{Tr}\{\rho |r(t)\rangle\langle s(t)|\}.$$

Using the equation of motion

$$\frac{\partial}{\partial t}(|r\rangle\langle s|) = i[H, |r\rangle\langle s|],$$

we obtain

$$\frac{\partial P_{rs}}{\partial t} = i\,\mathrm{Tr}\,\rho\left\{E_r|r\rangle\langle s| - E_s|r\rangle\langle s| + \sum_q V_{qr}|q\rangle\langle s| - \sum_q V_{sq}|r\rangle\langle q|\right\}.$$

Using the notation

$$E_{rs} = E_r - E_s,$$

and absorbing diagonal elements of V_{pq} into the E_s, so that

$$V_{rr} = 0 \text{ for all } r,$$

we have

$$\frac{\partial P_{rs}(t')}{\partial t} = i\left\{E_{rs} P_{rs}(t') + \sum_{q\neq r} V_{qr} P_{qs}(t') - \sum_{q\neq s} V_{sq} P_{rq}(t')\right\}.$$

We multiply by the integrating factor $\exp(-iE_{rs}t')$ and integrate from 0 to t which gives, for an initially diagonal state

$$P_{rs}(t) - P_{rr}(0)\,\delta_{rs} = i\int_0^t \exp[-iE_{rs}(t'-t)]\left(\sum_{q\neq r} V_{qr} P_{qs}(t') - \sum_{q\neq s} V_{sq} P_{rq}(t')\right) \mathrm{d}t'.$$

Iteration of this equation, with continuous withdrawal of the diagonal terms, gives the required result. Integration over a band of final-state energies, by

the same physical arguments as in the derivation of the "Golden-rule" equation for transition rates, gives a linear t-dependence, and indeed the resulting master equation is just the extension of the Golden-rule formula for the elements of the density matrix, when starting from a mixed state instead of a pure state. See also a recent paper by Pike and Swain.[16a]

We shall find that use of the master equation gives a unified approach to the problems of emission, absorption, generation of thermal light, and generation of laser light below, through, and above threshold, which I shall discuss in later sections.

2. The Detection of a Light Field

2.1 Emission and Absorption

One may consider the bound electron in the general theoretical situation described above, as acting as a detector of the field. To describe an absorption detector, which is the more usual kind, we withdraw the electron from contact with the heat bath, populate the lower state only, and then watch the development of its reduced density matrix σ as a function of time, under the action of the field. To be quite accurate, we should not allow the upper state to become appreciably populated, since emission would then take place back into the field and it would be partly an emission detector. We can therefore imagine the upper state to be greater than some ionisation energy, and that a mechanism is supplied which will withdraw the excited electron from the system. This process will constitute the "counting" of a photon. An emission detector can also be achieved both in theory and in practice, where now the upper state only is populated initially, and the effects of its depletion are measured. A He–Ne laser working at 0·6328 μm will conveniently detect infra-red radiation from a similar laser at 3·39 μm in this way. It will even put it out completely if it is sufficiently intense, and we have found this a convenient way of performing laser transient experiments. External damping of the radiation field is not essential for studying these processes.

The master eqn (9) of Section 1 very quickly gives the usual answers for the probabilities of emission and absorption. The matrix elements are:

$$V_{sr} = g\langle n_s\, b_s \mid b_u^{\dagger}\, b_l\, a + b_l^{\dagger}\, b_u\, a^{\dagger} \mid n_r\, b_r\rangle.$$

The first term only gives non-zero contributions when

$$n_s = n_r - 1, \quad b_s = b_u, \quad b_r = b_l$$

The value of this contribution will be $g\sqrt{n_r}$. The second term will give a contribution when

$$n_s = n_r+1, \quad b_s = b_l$$
$$b_r = b_u$$

with value $g\sqrt{(n_r+1)}$.

We first consider absorption. The simplest initial conditions at $t = 0$ are that the field is in state $|n\rangle$ and the electron in state $|b_l\rangle$; we wish to find the occupation probability of $|b_u\rangle$, that is, the value of the diagonal element σ_{uu}, after a time Δt. Summing over the radiation-field states we have, using $P_{rr}(0) = 0$,

$$\sigma_{uu}(\Delta t) = \sum_{n_r} P_{n_r b_u n_r b_u}(\Delta t) = \pi h_0 \Delta t \sum_{n_r, s} V_{rs} V_{sr} P_{ss}(0).$$

From the above calculation of the matrix elements V_{rs}, this reduces to

$$\sigma_{uu}(\Delta t) = \pi h_0 \Delta t \sum_{n_r} |V_{n_r b_u n_r+1 b_l}|^2 P_{n_r+1 b_l n_r+1 b_l}(0).$$

The only non-zero contribution in the sum over n_r occurs when

$$n_r+1 = n,$$

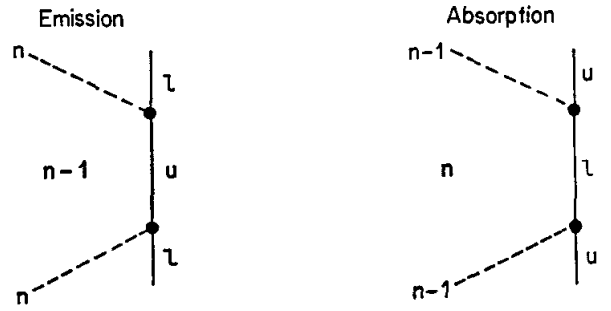
since this is the only state populated at $t = 0$. We have then finally

$$\sigma_{uu}(\Delta t) = \pi g^2 h_0 n \Delta t. \tag{10}$$

Similarly for emission

$$\sigma_{ll}(\Delta t) = \pi g^2 h_0 (n+1) \Delta t. \tag{11}$$

Figure 3 shows the contributions in diagrammatic form. All diagrams associated with master-equation perturbation theory are linked without further manipulation, and always begin and end with the same state.



$\sigma_{ll}(t) = \pi g^2 h_0 (n+1)t$ $\sigma_{uu}(t) = \pi g^2 h_0 nt$

FIG. 3. Diagrams of emission and absorption.

2.2 SINGLE COUNTING STATISTICS

Now an absorption detector placed in a beam of radiation consists of a very large number, N, of such absorbing electrons, in fact, so many that although the probability (eqn (10)) of each individual atom undergoing a transition is very small, nevertheless, the entire number of photons would be absorbed in a time of the order of $1/\pi g^2 h_0 nN$. However, the number of detecting atoms that can be used in practice is limited, firstly because the electrons in the upper state must be collectable and, secondly, the electrons must not decay back to the lower state, within the thickness of the detector. In practice taking into account the excited, but lost, electrons and the photons which pass unabsorbed through the detector, the ratio, η, of the number of successful "counts" to the number of incident photons is unfortunately no more than a few parts in a hundred (Foord *et al.*[17]). The photon-counting distribution $p(m, \Delta t)$, therefore, reproduces the photon-number distribution (assuming that the source, field and detector are in a steady state, and that photons absorbed in the detector are continuously replenished by the source) save for a random attenuation η.

Thus using Bernoulli's distribution (Scully[6])

$$p(m, \Delta t) = \sum_{n=m}^{\infty} \binom{n}{m} (\eta \Delta t)^m (1-\eta\, \Delta t)^{n-m}\, \rho_{nn}. \tag{12}$$

The most useful way to interpret theoretical calculations of ρ_{nn} by experiment is to construct the normalized factorial moments $n^{(r)}$ of the photon-counting distribution. These are defined as

$$n^{(r)} = \sum_m \frac{m!}{(m-r)!} p(m, \Delta t) \Big/ \left[\sum_m m p(m, \Delta t)\right]^r. \tag{13}$$

Using eqn (12) above

$$n^{(r)} = \sum_{nm} \frac{m!}{(m-r)!} \frac{n!}{m!\,(n-m)!} (\eta \Delta t)^m (1-\eta\, \Delta t)^{n-m}\, \rho_{nn} \Big/$$

$$\left[\sum_{nm} \frac{mn!\,(\eta\, \Delta t)^m}{m!\,(n-m)!} (1-\eta\, \Delta t)^{n-m} \rho_{nn}\right]^r \tag{14}$$

$$= \sum_n \frac{n!}{(n-r)!} \rho_{nn} \Big/ \left[\sum_n n \rho_{nn}\right]^r.$$

Thus the normalized factorial moments of the photon-counting distribution are identical with the normalized factorial moments of the photon-number

distribution. This remarkable result allows us to treat the photon-counting distribution as though it were exactly the photon-number distribution, independently of detector efficiency and beam attenuation, so long as interpretation is always made via the normalized factorial moments. Sometimes the normalizing denominator is omitted, in which case the function is denoted by $N^{(r)}$. (See also Peřina's chapter in this volume for further discussion.)

If the field has a coherent-state representation, the photon-counting distribution may be found by substituting for the diagonal element of the density matrix in the number representation (Glauber[18])

$$\rho_{nn} = \int \frac{P(\alpha)}{n!} |\alpha|^{2n} \exp(-|\alpha|^2)\, \mathrm{d}^2\alpha, \tag{15}$$

in eqn (12) above.

The result, after a little manipulation, is

$$p(m, \Delta t) = \int P(\alpha) \frac{(\eta\, \Delta t |\alpha|^2)^m}{m!} \exp(-\eta\, \Delta t |\alpha|^2)\, \mathrm{d}^2\alpha. \tag{16}$$

This formula was first derived by Mandel[19] in the classical case. For classical fields $|\alpha|^2$ $(= I)$ is the field intensity. For long sampling times the quantity $|\alpha|^2 \Delta t$ is replaced by

$$E(T) = \int_0^T \langle \alpha | a^\dagger a | \alpha \rangle\, \mathrm{d}t. \tag{17}$$

The matrix element in this integral can be a function of time; this requires a little discussion. The ensemble distribution ρ_{nn} can be converted to a stationary time distribution while still discussing the single-mode case, provided that the time scale of variations (coherence time) is long compared with the optical frequency. A spectrum analyser with a suitable Q factor would then show a very sharp line which varied in amplitude at different times. The mathematical description is the same as a Fourier analysis on an infinite time scale, which would give a description of the same field in terms of multiple modes:

$$\mathbf{E}^+(\mathbf{r}, t) = \sum_k (\tfrac{1}{2}\hbar\omega_k)^{\frac{1}{2}} u_k(\mathbf{r}) \exp(-i\omega_k t)\, a_k \text{ and c.c.} \tag{18}$$

In this description

$$E(T) = \int_0^T \langle \mathbf{E}^-(\mathbf{r}, t)\, \mathbf{E}^+(\mathbf{r}, t) \rangle\, \mathrm{d}t \tag{19}$$

The factorial moments can easily be expressed in terms of α by substituting eqn (16) in the definition, eqn (13). We obtain

$$n^{(r)} = \langle|\alpha|^{2r}\rangle/\langle|\alpha|^2\rangle^r. \tag{20}$$

The factorial moments of the photon-counting distribution are thus proportional to the actual moments of the "intensity-fluctuation distribution" $P(I)$. The same holds for "integrated" photon-counting distributions using eqn (19) above.

Discussions of $n^{(2)}$, the second factorial moment, or the closely related

$$\langle\Delta n^2\rangle/\langle n\rangle^2 \quad \left(= n^{(2)} + \frac{1-\langle n\rangle}{\langle n\rangle}\right),$$

played an important role in the early days of the subject. Its departure from unity, representing the value for shot noise, was then called excess noise and was the quantity usually measured. The first full photon-counting distributions reported with modern fast counting equipment were by Johnson *et al.*[20] at the Puerto Rico Quantum Electronics Conference in 1965. With such very accurate distributions, moments as high as $n^{(6)}$ have been successfully compared with theory, for thermal and laser sources (Jakeman *et al.*[21]) The first publication presenting moments higher than the second was by Freed and Haus.[22, 23]

Both the photon-counting distribution and its factorial moments may be derived from a single generating function $Q(s)$ (Glauber[4]). This is defined as the average over the photon-counting distribution

$$Q(s) = \sum_m (1-s)^m\, p(m, T). \tag{21}$$

A simple calculation shows that

$$p(m, T) = \frac{(-1)^m}{m!}\,\frac{d^m}{ds^m}\,Q(s)\Big|_{s=1} \tag{22}$$

$$N^{(r)} = (-1)^r \frac{\mathrm{d}^r}{\mathrm{d}s^r}\,Q(s)\Big|_{s=0} \tag{23}$$

and

$$Q(s) = \langle\exp(-\eta s E)\rangle. \tag{24}$$

The generating function is thus the Laplace transform of the intensity-

fluctuation distribution. It is also given directly by the relation

$$Q(s) = \sum_n (1-\eta s)^n \rho_{nn} \tag{25}$$

in the limit of very small T.

2.3 MULTIPLE COUNTING STATISTICS

If the transition probabilities of the detecting atoms are essentially independent of frequency over the range of frequencies used, the same type of arguments, as were outlined above, can be used to provide straightforward results for the multiple detection of photons in many modes, at a set of different space–time points x_n. The simplest experiment of this type is to measure the expectation value, $\langle m_1(x_1, \Delta x_1)\, m_2(x_2, \Delta x_2)\rangle$, for m_1 counts to be recorded over Δx_1 jointly with m_2 counts over Δx_2. If we make these intervals the same and small but non-overlapping, we find that

$$\langle m_1\, m_2\rangle = (\eta\, \Delta t)^2 \langle \mathbf{E}^-(x_1)\, \mathbf{E}^-(x_2)\, \mathbf{E}^+(x_2)\, \mathbf{E}^+(x_1)\rangle, \tag{26}$$

which we denote by

$$\langle m_1\, m_2\rangle = (\eta\, \Delta t)^2\, G^{(2)}(x_1, x_2). \tag{27}$$

If the intervals overlap completely, on the other hand, we have

$$\langle m_1{}^2\rangle = (\eta\, \Delta t)^2\, G^2(x_1, x_1) + \eta\, \Delta t\, G^{(1)}(x_1, x_1). \tag{28}$$

These formulae will be discussed in the last section.

The notation $G^{(r)}(x_1 \ldots x_{2r})$ was introduced by Glauber[24] for the rth-order correlation function of this type. The functions $G^{(r)}$ and $N^{(r)}$ are closely related (Glauber[4]). In fact, if the coordinates $x_1 \ldots x_{2r}$ are all the same, then the two functions are identical. Simplifying the expression (26) above, when x_1 and x_2 have the same space point and when the fluctuations of the field are stationary, we get the autocorrelation function of photon-counting fluctuations at a single detector

$$\begin{aligned} \langle m(\tau, \Delta t)\, m(0, \Delta t)\rangle &= (\eta\, \Delta t)^2\, G^{(2)}(\tau), && \tau > \Delta t \\ &= (\eta\, \Delta t)^2\, G^{(2)}(0) + \eta\, \Delta t\, G^{(1)}(0), && \tau = 0 \end{aligned} \tag{29}$$

This quantity may be measured directly and is of great importance in intensity-fluctuation spectroscopy of thermal light sources, as I shall explain in the next section. Its Fourier transform is also often measured and is called the intensity-fluctuation spectrum.

To conclude my discussion of the detection of optical fields, I shall make a few remarks concerning experimental apparatus and techniques. An ideal detector, in the sense described above, is essentially available in the modern

photomultiplier tube. Fortunately, an optical photon (~ 1 eV) is quite an energetic particle compared with room temperature ($\sim \frac{1}{40}$eV), and in the last few years, due largely to the work of Eberhardt[25] at I.T.T., optical detection has become virtually noiseless. Figure 4 shows the characteristics of such a tube. A large number of electrons ($\sim 10^7$) arrive at the anode of the tube when one photoelectron is released at the cathode, with a spread in this number which is closely predictable from the theory of cascade multiplication (Woodward[26]). The transit-time spread in this particular tube is large ($\sim 15\ \mu$s) but faster versions are available. Light fluxes of the order of one photon/min can be detected (Oliver and Pike[27]).

Most experimental work in photon statistics has been concerned with the measurement of $G^{(2)}(\tau)$ for spectroscopic purposes. In most cases this has been done simply by analysing the current output of the detector, using a scanning electrical filter (wave-analyser), to obtain the intensity–fluctuation spectrum. Less frequently a similar experiment in the time domain, that is, a scanning delay, has been used to measure this autocorrelation. This is the method of delayed coincidences. Although popular and simple, neither method is very efficient, since a parallel bank of filters, or delays, if feasible, would clearly give an extremely large gain. This extra information is avail-

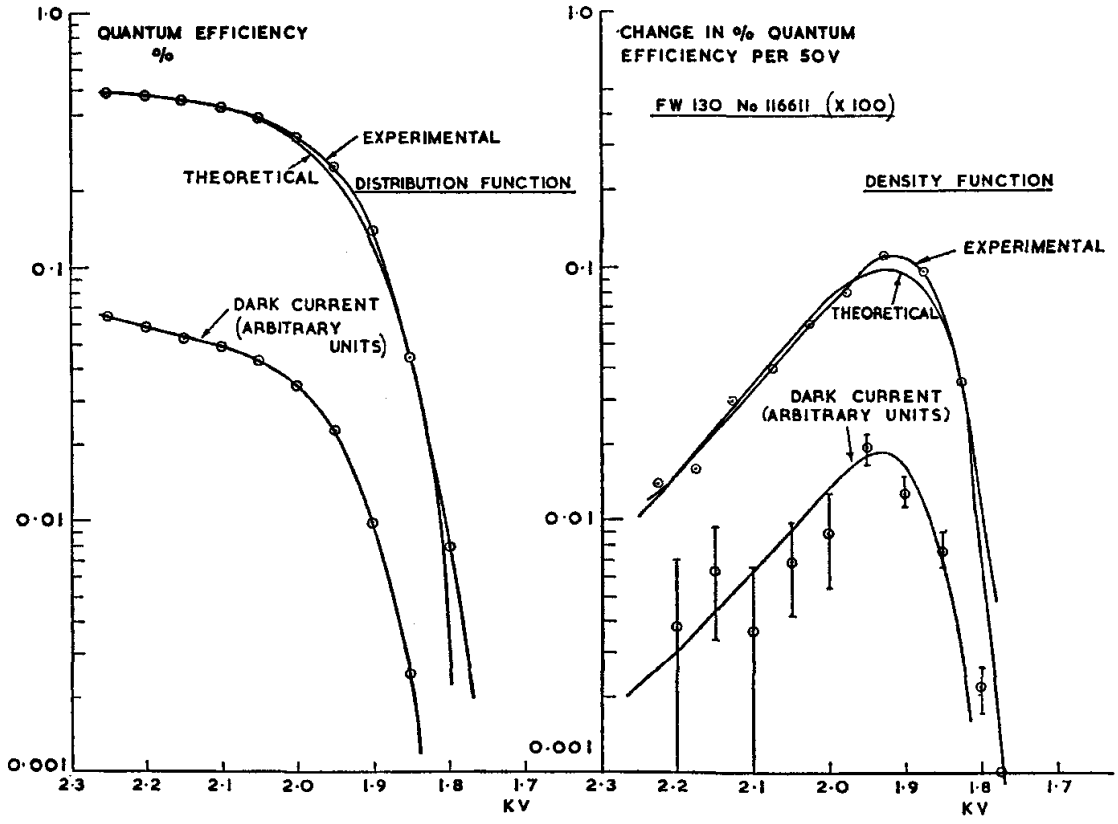


FIG. 4. Characteristics of the I.T.T. FW 130 photomultiplier tube. (After Foord *et al.*[17]).

able if a full correlation can be performed, that is, if a signal is progressively delayed and multiplied point by point with the original in a more or less continuous fashion. The difficulty here is the multiplication, which is normally too costly to perform. A great step forward, however, was made in the related microwave problem during the Second World War by van Vleck and his research assistant Middleton,[28] when they showed that the autocorrelation function of a "hard-limited" Gaussian signal was just $2/\pi$ times the arc sine of the original. Hard-limiting means giving the signal a unit amplitude when above a fixed "clipping level", usually the mean, and zero when below. Multiplication then involves only zeros and ones and can be much more easily performed.

In my own laboratory my colleagues and I have recently extended these methods to optical work. This has made intensity fluctuation measurements much easier and has greatly improved the accuracy of such measurements. The method which we have found most practical has been to call $m(0)$, 0 or 1, $(m_k(0))$, according to whether it is below or above an integer clipping level k, and to cross correlate this with the unclipped value $m(\tau)$. The analogue of the van Vleck–Middleton formula is then (Jakeman and Pike[29]) for thermal light

$$\frac{\langle m_k(0)\, m(\tau)\rangle}{\langle m_k\rangle \langle m\rangle} = 1 + \frac{1+k}{1+\bar{n}} |g^{(1)}(\tau)|^2 , \tag{30a}$$

where $g^{(1)}$ is a normalized form of $G^{(1)}(\tau)$, equal to $G^{(1)}(\tau)/G^{(1)}(0)$.

A simpler scheme is to clip at $k = 0$ and autocorrelate the clipped signal with itself. The result is[29]

$$\frac{\langle m_0(0) m_0(\tau)\rangle}{\langle m_0\rangle^2} = \left[1 + \frac{1-\bar{m}}{1+\bar{m}} |g^{(1)}(\tau)|^2\right] \left[1 - \frac{\bar{m}^2}{(1+\bar{m})^2} |g^{(1)}(\tau)|^2\right]. \tag{30b}$$

The above results are derived by making use of an extension of Glauber's generating function $Q(s)$ to two variables, by averaging over the joint distribution $p(m_1\ m_2)$. The function in this case is[29]

$$Q(ss') = \{\bar{n}^2 ss'[1 - |g^{(1)}(\tau)|^2] + \bar{n}(s+s') + 1\}^{-1}. \tag{30c}$$

In the sections below on laser statistics and applications, I shall give some examples of the use of clipping. $G^{(2)}(0)$ i.e. $N^{(2)}$, and higher factorial moments $N^{(r)}$ of photon-counting distributions have also been studied by a number of groups, including my own, and detailed considerations of statistical accuracy in periodic sampling of different types of light source have been made (Jakeman and Pike[30]). The basic point to emerge, as might be expected, is that the statistical accuracy in the determination of intensity fluctuations by photon-counting methods is limited by the ratio of the total time of the

experiment to the "coherence time" of the light, and not necessarily by the ultimate cycling speed of the sampling system. The hardware used for obtaining photon-counting distributions at the present time ranges from very expensive sampling and decoding electronic systems cycling at tens of megahertz, to simple methods utilising ordinary nuclear pulse-height analysers some three orders of magnitude slower. The original experiments of Freed and Haus[22] were a further three orders of magnitude slower. They used a mechanical tape punch to provide a store. Another few orders of magnitude takes us back to Rutherford and Geiger's study of the statistics of radio-active decay, performed in 1910.[31] They depressed a key by hand on to a moving paper ribbon to record counts of photons viewed by eye from a fluorescent screen. I shall give examples of measurements of factorial moments in the next section.

3. Special Fields

3.1 Photon statistics of thermal fields

The simple theory of the interaction between a single bound electron and a single-mode radiation field which I have given above will again suffice, taken to first order only in the interaction, for the discussion of thermally generated light fields. In this case, the electron is the generator of photons instead of being the detector, as in the application in the previous section.

The master equation reads as above (eqn (9)) but the populations σ_{uu} and σ_{ll} are now taken to be in thermal equilibrium at a positive temperature T, that is,

$$\frac{\sigma_{uu}}{\sigma_{ll}} = \exp(-\hbar\omega/kT). \tag{31}$$

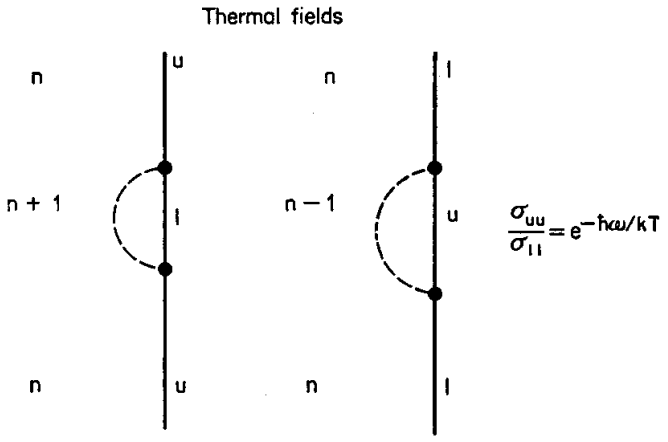


Fig. 5. Diagrams for thermal-light generation.

The two diagrams of Fig. 5 now give the only contributions and we have, tracing this time over the atomic coordinates, and summing over the number of atoms, N,

$$\rho_{nn}(t) = \rho_{nn}(0) + \pi g^2 h_0 N t\,[(n+1)\,(\rho_{n+1,\,n+1}\,\sigma_{ll} - \rho_{nn}\,\sigma_{uu}) + n(\rho_{n-1,\,n-1}\,\sigma_{uu} - \rho_{nn}\,\sigma_{ll})]. \quad (32)$$

A similar equation and its generalisation, which we shall introduce below for the laser problem, has formed the basis of much of the work of Lamb and Scully, who derived it by the Liouville–Neumann iteration for the density operator, but without performing the energy integrals to give the linear t-dependence.

The equation may be written in the "adiabatic" approximation in which the atomic populations can be considered as unchanging.

$$\frac{\mathrm{d}\rho_{nn}}{\mathrm{d}t} = -A(n+1)\,\rho_{nn} + C(n+1)\,\rho_{n+1,\,n+1} - Cn\rho_{nn} + An\rho_{n-1,\,n-1}, \quad (33)$$

where

$$A = \pi g^2 h_0\, N\sigma_{uu} \quad (34)$$

and

$$C = \pi g^2 h_0\, N\sigma_{ll}. \quad (35)$$

We look for solutions of the form

$$\rho_{nn} = \sum_m \rho_{nn}^m \exp(-\lambda_m t). \quad (36)$$

For finite, non-zero solutions,

$$\lambda_0 = 0 \quad (37)$$

and

$$\lambda_n = (C-A)\,n. \quad (38)$$

The solution is, when no photons are present at $t=0$,

$$\rho_{nn}(t) = \frac{C-A}{C-A\exp[-(C-A)t]}\left(\frac{A(1-\exp[-(C-A)t]}{C-A\exp[-(C-A)t]}\right)^n. \quad (39)$$

The time dependence shows how the distribution decays to the stationary state. $\lambda_1 (= \pi g^2 h_0 N(\sigma_{ll} - \sigma_{uu})$ is essentially the Wigner–Weisskopf inverse linewidth.

In terms of the mean value of the occupation number, $\bar{n}$, the time-independent solution is the familiar geometric distribution

$$\rho_{nn} = \frac{\bar{n}^n}{(\bar{n}+1)^{n+1}}, \tag{40}$$

where we have calculated that

$$\bar{n} = \sum_{n=0}^{\infty} n\rho_{nn} = \frac{1}{\exp(\hbar\omega/kT)-1}. \tag{41}$$

This, of course, is known as the Bose factor. The same distribution seems to result in all orders of perturbation theory. The generating function for this distribution is

$$Q(s) = \frac{1}{1+\eta\Delta t\,\bar{n}s}. \tag{42}$$

The factorial moments are

$$N^{(r)} = (\eta\Delta t\,\bar{n})^r\, r!. \tag{43}$$

The photon-counting distribution is

$$p(m, \Delta t) = \frac{(\eta\Delta t\,\bar{n})^n}{(\eta\Delta t\,\bar{n}+1)^{n+1}}. \tag{44}$$

The field has P-representation with the intensity–fluctuation distribution

$$P(|\alpha|^2) = \frac{1}{\pi\langle|\alpha|^2\rangle}\exp(-|\alpha|^2/\langle|\alpha|^2\rangle). \tag{45}$$

I should not need to emphasise that the fact that a field has a positive and well-behaved P-representation, such as the Gaussian one above, does not imply that it is a classical field (Glauber[18]). This is clearly shown by the thermal field above, where the factor unity representing spontaneous emission (quantum noise) was essential for the calculation. The point has been the subject of some controversy (Mandel and Wolf[32]).

Examples of light fields having the same statistical properties as thermal fields can, in fact, be generated by imposing a Gaussian amplitude modulation on a coherent laser beam by passing it through a rotating ground-glass screen. These are so-called "quasi-thermal" sources, other examples of which I shall discuss more fully in the later section on scattered laser light.

Intensity fluctuations, showing the Bose character, were obtained a number of years ago in this way.[33, 34]

Figure 6 shows an early result from R.R.E. which appears in Professor Kastler's revision of Bruhat's text "Thermodynamique". Here quasi-thermal and laser photon-counting distributions are directly compared on an oscillogram.

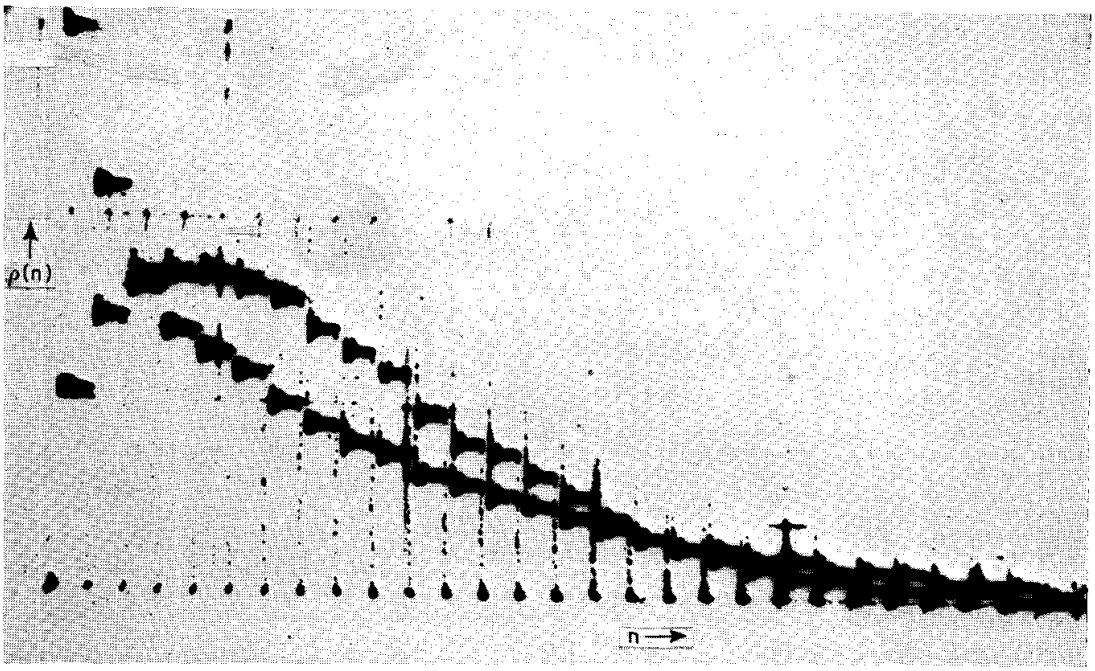


FIG. 6. Oscillogram of photon-counting distributions of quasi-thermal and laser light. (Reproduced from Bruhat "Thermodynamique," Masson, Paris, 1968)

A "single-mode" thermal field can vary in time as discussed above; the amplitude-modulation side bands which result, form an optical spectrum. On an intermediate time scale of Fourier analysis many adjacent modes, each varying in amplitude, can be considered. The field will also vary in space. The correlation functions $G^{(r)}$ introduced by Glauber can be used to characterise the statistical properties of a general superposition of thermal modes. The $N^{(r)}$ derived above are the values of $G^{(r)}$ for zero coordinate differences.

The optical spectrum, for instance, of a stationary field is defined as the Fourier transform of the autocorrelation function mentioned above,

$$G^{(1)}(\mathbf{r}, \tau) = \langle \mathbf{E}^-(\mathbf{r}, \tau)\, \mathbf{E}^+(\mathbf{r}, 0) \rangle. \tag{46}$$

The intensity–fluctuation spectrum is the Fourier transform of

$$G^{(2)}(\mathbf{r}, \tau) = \langle \mathbf{E}^-(\mathbf{r}, \tau)\, \mathbf{E}^-(\mathbf{r}, 0)\, \mathbf{E}^+(\mathbf{r}, 0)\, \mathbf{E}^+(\mathbf{r}, \tau) \rangle. \tag{47}$$

On the other hand, spatial correlations may be investigated at a fixed time; for instance, by measuring the function

$$G^{(2)}(\mathbf{r}_1\,\mathbf{r}_2, 0) = \langle \mathbf{E}^-(\mathbf{r}_1, 0)\,\mathbf{E}^-(\mathbf{r}_2, 0)\,\mathbf{E}^+(\mathbf{r}_2, 0)\,\mathbf{E}^+(\mathbf{r}_1, 0)\rangle. \tag{48}$$

The full range of arbitrary rth-order correlations are, in principle, open to measurement. The values of and relations between such correlation functions are the basic problems of photon statistics. For thermal (Gaussian) fields it can be shown[35, 18] that

$$G^{(2)}(\mathbf{r}, \tau) = [G^{(1)}(\mathbf{r}, 0)]^2 + |G^{(1)}(\mathbf{r}, \tau)|^2 \tag{49}$$

and further that[18]

$$G^{(r)}(x_1 \ldots x_2) = \sum_{\text{Perms}} \prod_{j=1}^{r} G^{(1)}(x_j, x_{p(r+j)}). \tag{50}$$

The rth-order correlation function for Gaussian fields is a symmetrical sum of products of first-order correlation functions.

The interpretation of the $G^{(r)}$ as multiple photon-detection probabilities shows that such factorization properties imply some degree of independence of photon-detections at different space–time points. If all such events were independent, the field would be completely coherent (lack of knowledge of the photon number implies gain of information concerning the conjugate phase variable). The highest degree of factorization possible for a thermal field is[36]

$$G^{(r)}(x_1 \ldots x_{2r}) = r! \prod_{j=1}^{r} \mathscr{E}(x_j) \prod_{j=r+1}^{2r} \mathscr{E}(x_j). \tag{51}$$

Such a field has been called ideally incoherent, first-order coherent, or weakly coherent.[37]

This factorization has been confirmed experimentally up to $G^{(6)}$ for a chaotic "quasi-thermal" field produced by scattering a laser beam from polystyrene balls undergoing Brownian motion in water.[21] These results are shown, together with the results for an ideal laser, in Table I. The factorization of the joint photon-counting probability $G^{(2)}$ given by eqn (49) permits a symmetrical optical spectrum to be found by correlation of photon-counting fluctuations, using eqn (29) above. This is the basic theory of the experiments of Forrester *et al.*,[1] and Hanbury-Brown and Twiss.[2] We have, at a single point

$$\langle m(\tau, \Delta t)\, m(0, \Delta t)\rangle = (\eta \Delta t)^2 \{[G^{(1)}(0)]^2 + |G^{(1)}(\tau)|^2. \tag{52}$$

TABLE I

Normalized factorial moments for coherent source[a]

(After Jakeman *et al.*[21])

Normalized factorial moment	10^7 samples Experiment	Theory
$n^{(1)}$	1·0000	1·0000
$n^{(2)}$	1·0002	1·0000±0·0009
$n^{(3)}$	1·0001	1·0000±0·0017
$n^{(4)}$	1·0006	1·0000±0·0030
$n^{(5)}$	1·003	1·000±0·012
$n^{(6)}$	0·994	1·000±0·036

Normalized factorial moments for incoherent source

Normalized factorial moment	10^5 samples Experiment	Theory	9×10^6 samples Experiment	Theory
$n^{(1)}$	1·000±0·036	1·000±0.038	1·000	1·000
$n^{(2)}$	2·00±0·15	2·00±0·16	2·004	2·000±0·017
$n^{(3)}$	6·05±0·85	6·00±0·85	6·05	6·00±0·09
$n^{(4)}$	24·7±5·9	24.0±5·7	24·7	24·0±0·6
$n^{(5)}$	130±45	120±49	130	120±5
$n^{(6)}$	805±461	720±510	805	720±54

[a] Similar results were presented at the Conference CI07 in Paris, May 1966, but have not previously been published.

The requirements on the extensions in space and time over which the above formula is valid are known as coherence conditions, and are simply derived in any experimental situation with thermal light, by geometrical and spectral parameters. The factorization is, of course, used conversely to determine these parameters, by varying the spatial size and sampling time of the detector.

The effect of changing the interval Δt to a finite value T, comparable with the coherence time of a Gaussian source with Lorentzian spectrum of half-width Γ $(= 1/T_c)$, is given by Jakeman and Pike, (unpublished)

$$g^{(2)}(kT, T) = 1 + \frac{(\cosh \gamma - 1)}{2\gamma^2} \exp(-2k\gamma), \text{ for } k > 0 \tag{53}$$

$$g^{(2)}(0, T) = n^{(2)} = 1 + \gamma - \tfrac{1}{2}\gamma^2 + \frac{\exp(-2\gamma)}{2\gamma^2}, \text{ for } k = 0, \tag{54}$$

where $\gamma = T/T_c$, and the small $g^{(r)}$ is a normalized version of $G^{(r)}$, formed by

dividing by $[G^{(1)}]^r$. These results are shown in Fig. 7. The time scale on the figure is for scattering of laser light from a protein molecule, but the form of the curves holds for any chaotic source with Lorentzian spectrum. The normalized second factorial moment $n^{(2)}$ tends to two as the sampling time is reduced to zero, and to one for infinite sampling time.

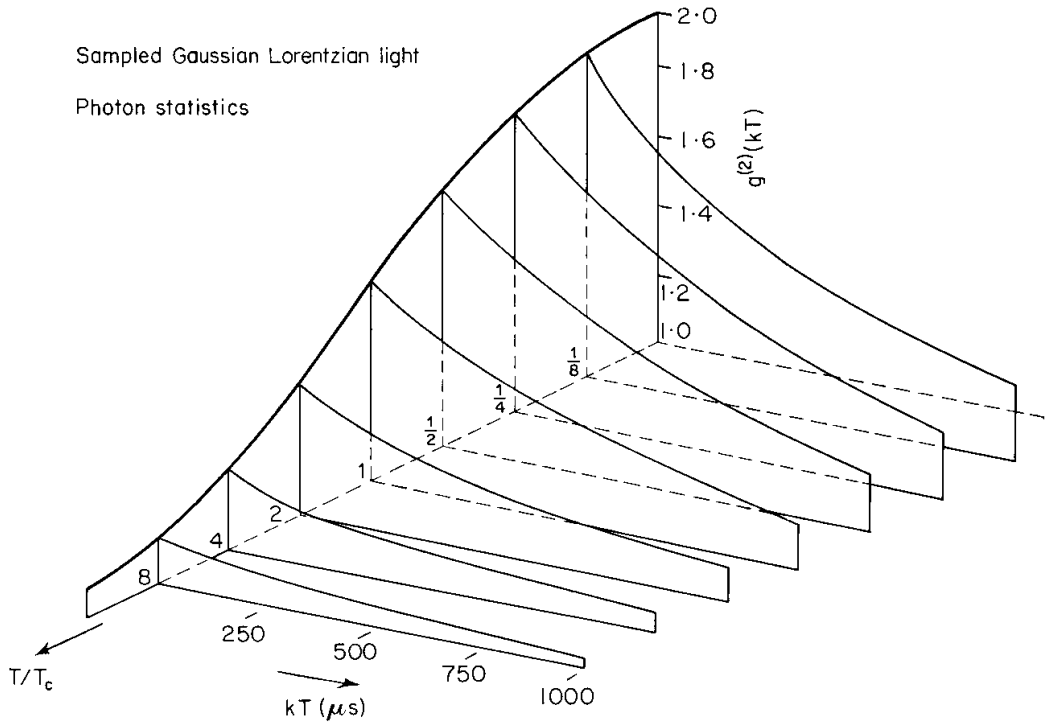


FIG. 7. The normalized second-order correlation function, $g^{(2)}(\tau)$, $(\tau = kT)$, for integrated thermal light.

The effects of finite sampling time on the photon-counting distribution, which one can see in $n^{(2)}$ in Fig. 7, have been discussed in a semiclassical formalism in the limit of large T by Mandel,[38] and more recently by Glauber,[4] Bédard[39] and very fully by Jakeman and Pike.[40]

The fluctuations are averaged out as the sampling time T reaches and exceeds the coherence time T_c. This effect may be used to measure the width of a spectrum of known shape.[41]

3.2 PHOTON STATISTICS OF LASER FIELDS

There have been a very large number of publications on this subject. The approaches used may be classified into three groups:

(1) use of a Langevin-type equation;

(2) use of a quantum mechanical Fokker–Planck equation;

(3) use of the equation of motion for the density matrix (master equation).

Clearly I have not time to go into detail in reviewing all these papers. (A review by Lax[7] of his own numerous papers ran to three hundred pages in a recent Brandeis University summer school!) I shall try to pick out, however, the essence of all this work as it applies to actual statistical experiments on laser light. Well below its threshold, the laser is the same as a thermal source, and its photon-number distribution is the geometric distribution; $n^{(2)}$ takes the value two, due to the typical fluctuations discussed above of a field in equilibrium with a thermal reservoir. This would hold independently of temperature for all positive temperatures. Well above threshold, however, a laser produces Poisson photon-counting statistics; $n^{(2)}$ takes the value unity and the fluctuations are damped out by the saturation mechanism of the oscillation. Between these two limits, the statistical properties of the steady state, in particular, the behaviour of $n^{(2)}$ and ρ_{nn}, have been given by Risken,[42] by Lax and Louisell[43] and may also be found in Scully, Lamb and Stephen.[44] The statistical properties of the non-steady state have also been widely studied by the same workers and others.

The effects can again be predicted using the simplified theoretical model I have already used twice above. This time the master equation has to be written to higher order and the effects of the loss of photons from each state $|n\rangle$ at a rate Cn must be taken into account. In the simplest case the (pumping) reservoir is assumed to depopulate completely the electron lower state. Thus at $t = 0$, $\sigma_{ll} = 0$. The theory is not much more difficult if a finite occupation of the lower state is allowed but this does not add any significant extra physics.

The master equation for the time development, with N atoms in the field, is

$$\begin{aligned} P_{rr}(t) = P_{rr}(0) + \pi h_0 N t \sum_s \{ & [V_{rs} V_{sr} \\ & + \tfrac{1}{8}(\pi h_0)^2 \sum_{s_1 s_2} (-4 V_{rs_1} V_{s_1 s_2} V_{s_2 s} V_{sr} + 6 V_{rs_1} V_{s_1 s} V_{ss_2} V_{s_2 r} \\ & - 4 V_{rs} V_{ss_1} V_{s_1 s_2} V_{s_2 r})] [P_{ss}(0) - P_{rr}(0)] + \ldots \} \\ & - C t n_r \rho_{n_r n_r} + C t (n_r + 1) \rho_{n_r + 1,\, n_r + 1}. \end{aligned} \tag{55}$$

The last term, representing a loss from the $n_r + 1$ level, is equivalent to a gain in the n_rth level. The same two terms could be arrived at by postulating absorbing atoms, but the lack of non-linear terms is then embarrassing. The diagrams which give contributions to the fourth-order terms are shown in Fig. 8. These are easily evaluated, and although these alone would give a

satisfactory model for lasing action over a wide range of operation, a slightly more satisfactory result is obtained when the complete series of higher-order terms is summed. The diagrams required are simple generalizations of those in Fig. 8, and the final result when traced over the electronic levels is the equation

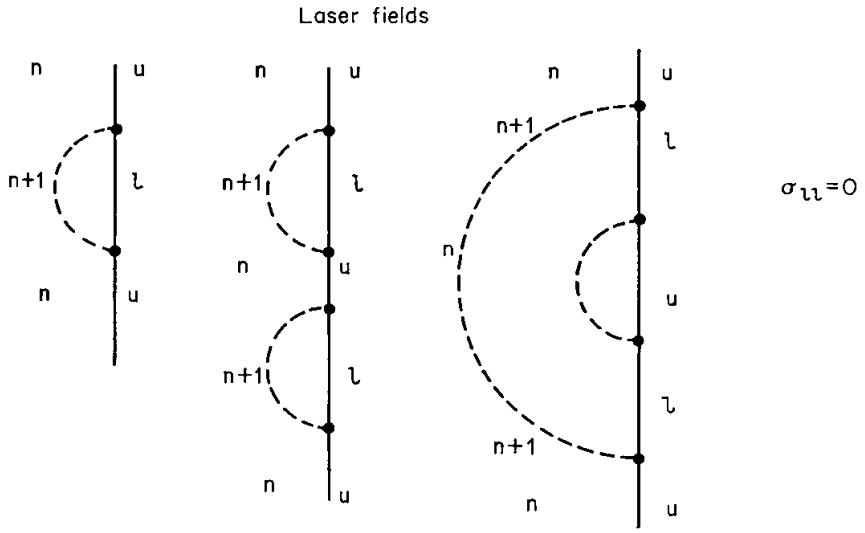


FIG. 8. Diagrams for fourth-order atom-field interaction.

$$\frac{d\rho_{nn}}{dt} = -A(n+1)\left[1+(n+1)\frac{B}{A}\right]^{-1}\rho_{nn}$$

$$+An\left[1+n\frac{B}{A}\right]^{-1}\rho_{n-1,\,n-1}-Cn\rho_{nn}+C(n+1)\,\rho_{n+1,\,n+1}, \tag{56}$$

where

$$B = (\pi g h_0)^2 A. \tag{57}$$

The equation is due to Scully and Lamb.[45]

We again look for solutions of the form

$$\rho_{nn} = \sum_m \rho_{nn}^m \exp(-\lambda_m t). \tag{58}$$

The stationary-state solution corresponds to $\lambda_0 = 0$. It is[45]

$$\rho_{nn} = Z^{-1}\prod_{k=0}^{n}\frac{A}{C}\left[1+\frac{B}{A}k\right]^{-1}, \tag{59}$$

Z being the normalization constant. Further evaluation gives

$$\rho_{nn} = \frac{(A^2/BC)^n\,(A/B)!}{[n+(A/B)]!\,[{}_1F_1\,(1;\,(A/B)+1,\,A^2/BC)]}. \tag{60}$$

In the α-representation an exactly equivalent form is given by[46]

$$P(|\alpha|^2) = Z^{-1}\exp(-|\alpha|^2)\,(\langle|\alpha|^2\rangle - |\alpha|^2)^{(\langle|\alpha|^2\rangle - C)} \tag{61}$$

and the first term of an expansion of this, about the mean value, gives the expression

$$P(|\alpha|^2) = Z^{-1}\exp\left(-\frac{|\alpha|^4}{4} + a\frac{|\alpha|^2}{2}\right) \tag{62}$$

found by Risken,[42] Fleck,[47] Hempstead and Lax[48] and Lax and Louisell.[43] This distribution has the desired properties of approaching the geometric distribution as $A/C \to 0$ and the Poisson distribution as $A/C \to \infty$.

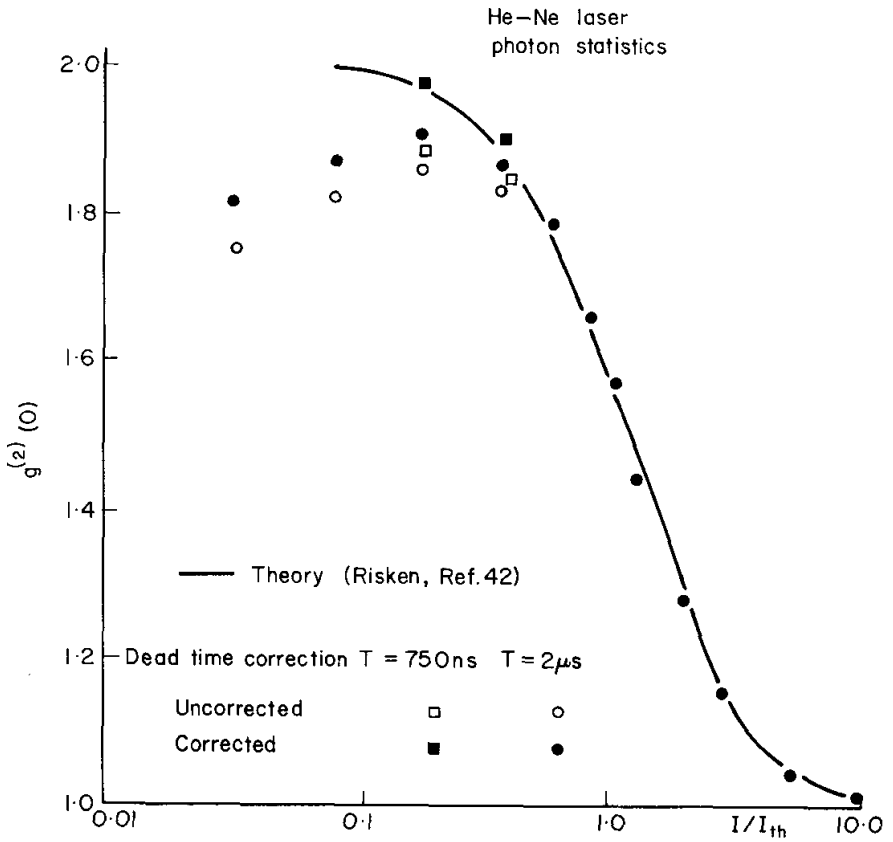


FIG. 9. The normalized second-order factorial moment, $n^{(2)}$ $(= g^{(2)}(0))$, of the photon-number distribution of a single-mode gas laser as a function of output power, showing the effect of resolving time corrections and finite sampling time.

Its lower normalized factorial moments have been evaluated to compare with experimental photon-counting results, and good agreement is found. The first experiments of this nature were by Freed and Haus[22] and by Armstrong and Smith,[49] where $n^{(2)}$ was compared with theory. The development of highly stable single-mode lasers has made such measurements now very easy, and much more accurate comparisons have been made by Professor Arecchi's group[50] in Milan, Professor Korenman[51] and his colleagues at the University of Maryland and my own group at R.R.E.[52a] Instrumental effects become important at the few percent accuracy level and corrections must be made for the finite ratio of detector resolving time to sampling time.[52b] This is shown in Fig. 9 where I have shown both the raw and corrected data for $n^{(2)}$ using a Spectra-Physics model 119 laser. For sampling time T as large as 2 μs a theory of integrated laser fluctuations is required. This problem has been treated by Lax and Zwanziger.[76] Experiments with several detectors have been performed by Davidson and Mandel.[53]

The higher eigenvalues λ_m and the corresponding ρ_{nn}^m have been calculated by Risken and Vollmer[54] and by Lax and Louisell,[43] and thus the value of $G^{(2)}(\mathbf{r}, \tau)$ can be compared with theory. In Fig. 10 is shown a three-dimensional picture of this function as the laser varies from well below, to well above

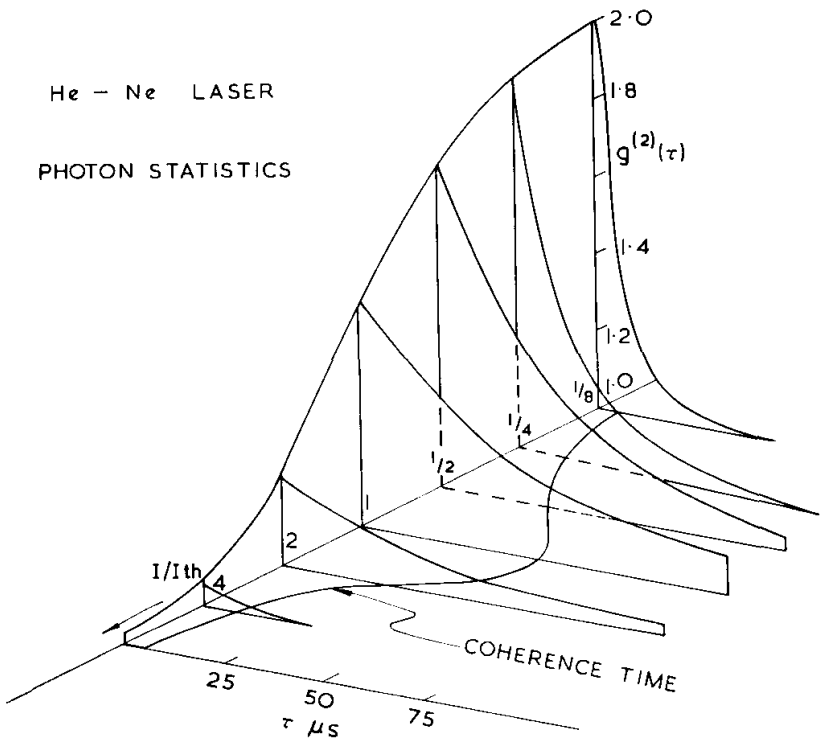


FIG. 10. The normalized second-order correlation function, $g^{(2)}(\tau)$ for a single-mode gas laser.

threshold. The curves were constructed from experimental measurements obtained at R.R.E., by the method of autocorrelation of clipped photon-counting fluctuations mentioned above.† In Fig. 11 are shown the corresponding intensity-fluctuation spectra using an ordinary wave analyser.

The calculations of Risken and Vollmer[54] predict that $G^{(2)}(\tau)$ should consist of a dominant single exponential below threshold, but that appreciable fractions of several faster exponential decays contribute in the region immediately above threshold. McCumber[55] and Mezrich,[56] however, predict essentially a single exponential decay in the threshold region. All theories give the "line-narrowing" at threshold, characteristic of second-order phase transitions. The role of order-parameter is played by $2-n^{(2)}$,

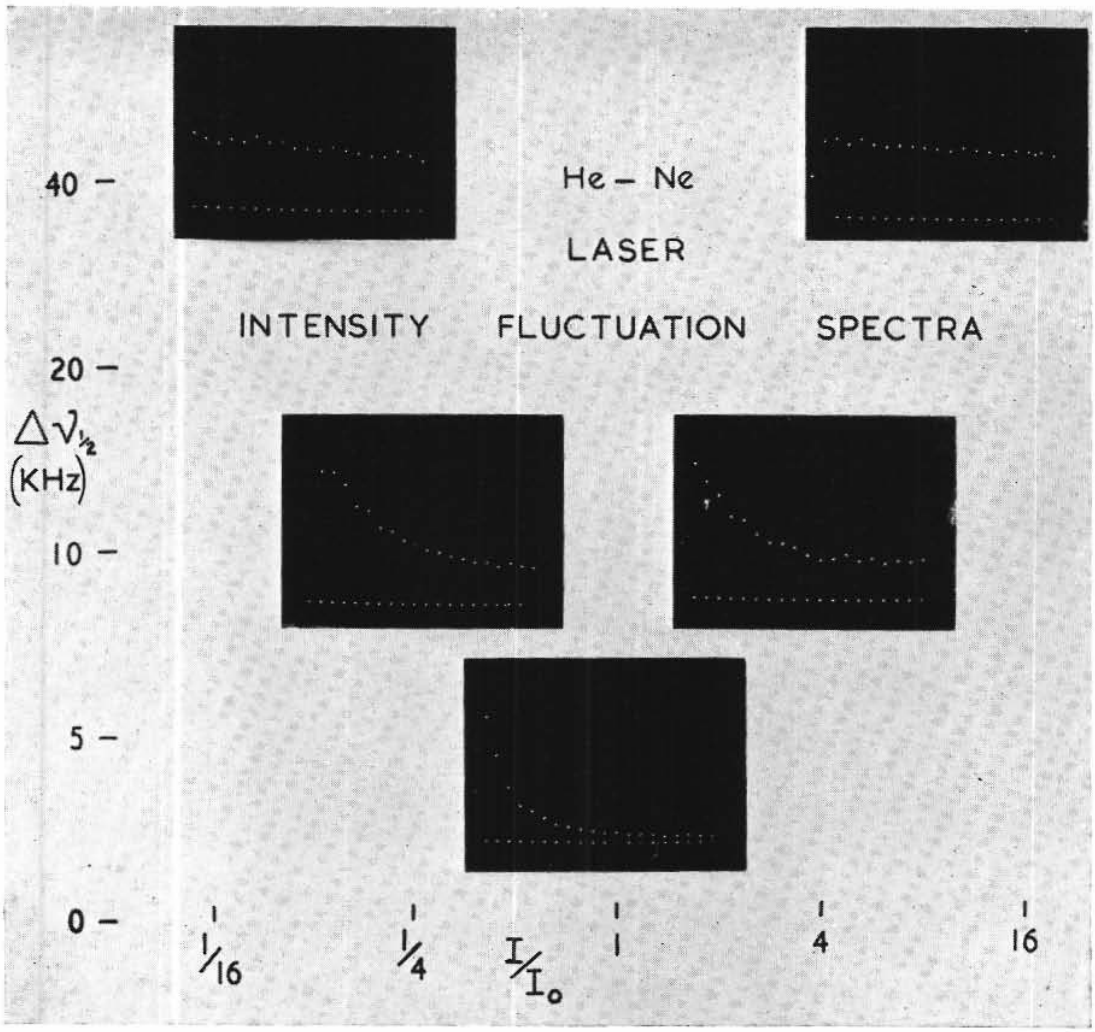


FIG. 11. The intensity-fluctuation spectrum of a single-mode gas laser near threshold. The traces shown correspond to the coordinates at their centres.

† The method is valid for arbitrary statistics at low count rates.

which is unity in the ordered (lasing) state and zero in the disordered (thermal) state. It has been realized for some time in the study of phase transitions and in transport theory, that the simple assumption for a correlation function, made almost universally, viz.

$$\langle \alpha(\tau)\, \alpha^*(0) \rangle = \langle |\alpha(0)|^2 \rangle \exp(-\lambda t) \tag{63}$$

may be unrealistic, since many different decay rates may contribute. Here in the laser, we have a very attractive system for detailed investigation. Arecchi *et al.*[57] have made spectral measurements which support the analysis of Risken and Vollmer. In my own group, in addition to spectral analysis, we have employed the new methods explained above for obtaining the correlation function directly. In Fig. 12 is shown an autocorrelation

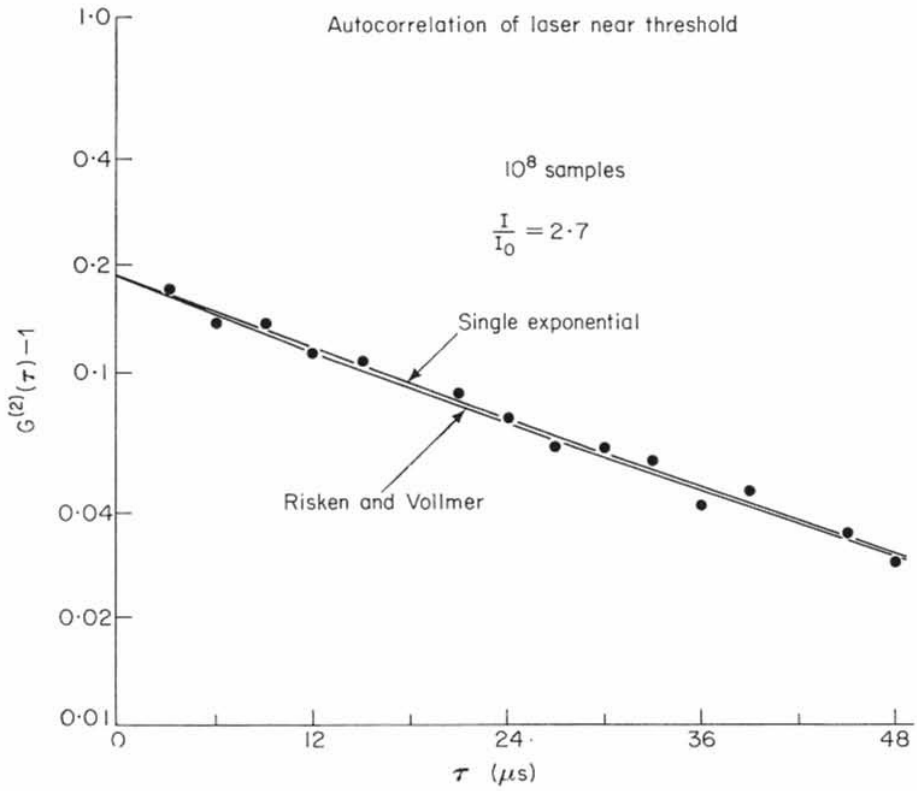


FIG. 12. The autocorrelation function $g^{(2)}(\tau)$ of the photon-counting fluctuations of aa single-mode gas laser at 2·7 times threshold output power, measured by the method of single-clipped autocorrelation (SCAC).

at 2·7 times threshold power which is in the critical region to test the theory. It is quite an accurate measurement, some 10^8 delays having been sampled. Preliminary analysis of such results shows that a valid confirmation of Risken and Vollmer's results will be difficult, since the departure from a single exponential decay is extremely small. With regard to experimental

technique, it is necessary to perform photon-counting experiments simultaneously with a spectral analysis of intensity fluctuations, to fix the threshold position absolutely. This is not necessary when investigating the autocorrelation, since $G^{(2)}(0)$ immediately gives the position with respect to threshold.

The experiments I wish to mention finally in this section on laser statistics are laser transients. The equation of motion for ρ_{nn} has been integrated numerically for transient conditions by Scully,[6] and the results show how the laser oscillations build up from noise below threshold. Arecchi *et al.*[50] have performed such experiments by Q-switching a laser with a Kerr cell inside the cavity, and have successfully compared the results with the theory. The results are shown in Fig. 13.

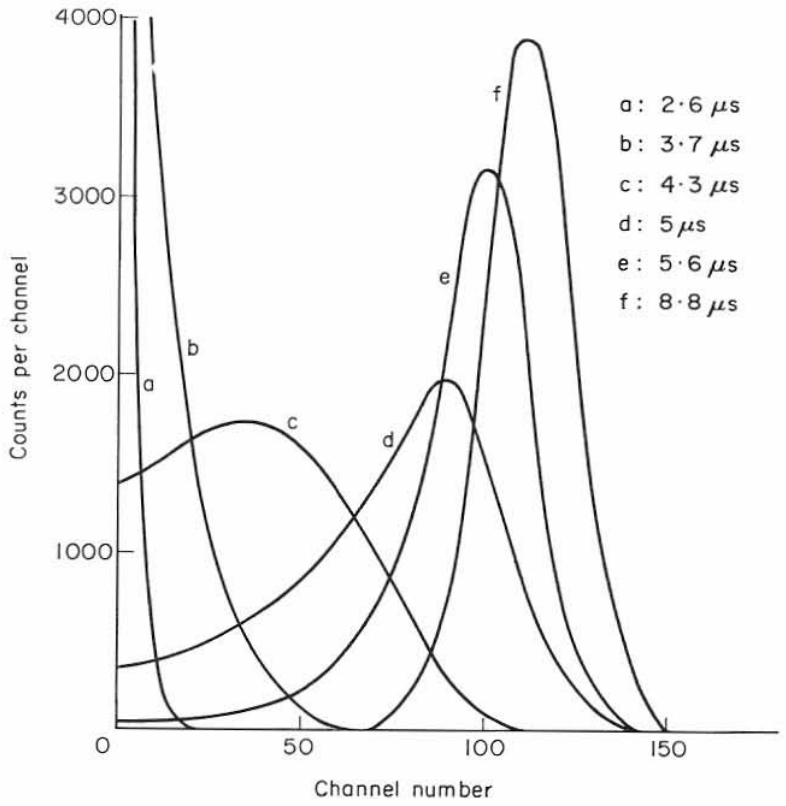


FIG. 13. Photon-counting distributions of a single-mode gas laser in transient operating conditions (After Arecchi *et al.*[50]).

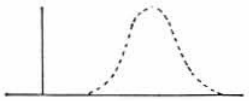
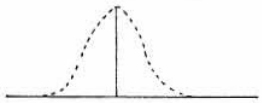
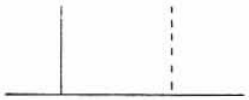
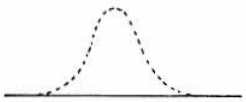
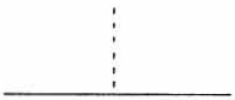
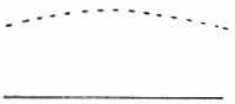
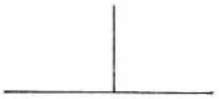
3.3 PHOTON STATISTICS OF MIXED FIELDS

Having discussed the case of thermal fields and laser fields, we come now to the consideration of superpositions of the two types. We take the laser

to be operating well above its threshold, so that its counting statistics are Poisson. It then acts as a "local oscillator" against which the thermal field can be heterodyned. Many workers have studied the statistics of such fields and in Table II is given a bibliography showing the results obtained to date.

TABLE II

Photon counting statistics of Gaussian–Lorentzian and coherent fields—a bibliography (After Jakeman and Pike.[58])

Field	Limit	$Q(s)$	$P(E)$	$p(n, T)$	$N^{(2)}$	$N^{(r)}$
		present paper			present paper	
	$\omega_q = \omega_0$	present paper			present paper	
	$\Gamma \to 0$	present paper	present paper		present paper	
	$\Gamma \to \infty$ $\omega_q = \omega_0$	present paper	present paper	present paper	present paper	present paper
	$\Gamma \to 0$ $\omega_q = \omega_0$	*G*66	*R*45[a] *P*67	*G*66 *L*65	*L*65	*G*45[a] *G*66 *L*65[d]
	$\langle E_c \rangle = 0$	*H*64	*J*68a[e]	*B*66[d] *J*68a[e]	*B*66[e] *J*68a[e]	*B*66[e]
	$\langle E_c \rangle = 0$ $\Gamma = 0$	*G*65	*R*45[a] *M*64	*M*59	*R*45[a] *M*58	*R*45[a] *G*66 *L*65[d]
	$\langle E_c \rangle = 0$ $\Gamma \to \infty$	*G*65[b] *H*64[b]	*J*68a	*M*59[b] *MP*65 *J*68b[f]	*Pu*56[c] *M*58[c] *G*65	*G*65
	$\langle E_i \rangle = 0$	*G*65	*R*45[a] *M*64	*G*63 *M*64	*R*45[a] *G*65	*R*45[a] *G*65

- - - - Incoherent field E_i.
——— Coherent field E_c.

a Mathematically equivalent calculation.
b Not formal expansions in $1/\gamma$.
c In these early papers bunching was thought to be a property of photons.
d Recurrence relations only.

e Calculated numerically.
f To second order in $1/\gamma$.
G Glauber, R. J., *Phys. Rev. Lett.* **10,** 84 (1963); *In* "Quantum Optics and Electronics" (eds. C. de Witt *et al.*), p. 65. Gordon and Breach, New York (1965); *In* "Physics of Quantum Electronics" (eds. P. L. Kelly *et al.*) p. 788. McGraw–Hill, New York (1966).
R Rice, S. O. *Bell. Syst. tech. J.* **24,** 46 (1945).
L Lachs, G. *Phys. Rev.* **138,** B1012 (1965).
H Helstrom, C., *Proc. Phys. Soc.* **83,** 777 (1964).
J Jakeman, E. and Pike, E. R., (a) *J. Phys. A.* **1,** 128 (1968); (b) *J. Phys. A.* **1,** 627 (1968).
B Bédard, G., *Phys. Rev.*, **151,** 1038 (1966).
M Mandel, L., *Proc. Phys. Soc.*, **72,** 1037 (1958); *Proc. Phys. Soc.* **74,** 233 (1959); Proceedings of the 3rd Conference on Quantum Electronics, Columbia University Press, New York (1964).
Pu Purcell, E. M., *Nature, Lond.*, **178,** 1449 (1956).
MP McLean, T. P. and Pike, E. R., *Phys. Lett.* **15,** 318 (1965).
P Peřina, J., *Phys. Lett.* **24A,** 333 (1967).

The table also includes the limiting single-field cases for reference. This table is taken from a recent paper on the statistics of heterodyne detection (Jakeman and Pike[58]). For the general case where the laser frequency, ω_q, differs from the centre of the thermal-field spectrum, ω_0, and where this spectrum is Lorentzian and of arbitrary half-width Γ $(= \gamma/T)$, only the generating function and the second factorial moment of the counting distribution have been obtained. The latter has the value

$$N^{(2)} = \langle n_i \rangle^2 \left(1 + \frac{1}{\gamma} - \frac{1}{2\gamma^2} + \frac{\exp(-2\gamma)}{2\gamma^2}\right) + 2\langle n_i \rangle \langle n_c \rangle \times \left[1 + \frac{2\exp(-\gamma)\{(\gamma^2 - \Omega^2)\cos\Omega - 2\gamma\Omega\sin\Omega\}}{(\Omega^2+\gamma^2)^2} + \frac{2\gamma}{\Omega^2+\gamma^2} + \frac{2(\Omega^2-\gamma^2)}{(\Omega^2+\gamma^2)^2}\right] + \langle n_c \rangle^2 \tag{64}$$

where n_i is the incoherent and n_c the coherent signal and

$$\Omega = (\omega_q - \omega_0)T. \tag{65}$$

The behaviour as a function of γ is shown in Fig. 14 for the particular parameters given. Of more practical interest is the autocorrelation function or the intensity-fluctuation spectrum of a mixed field. It can be shown that under reasonable conditions the complete optical spectrum of a source with arbitrary statistical properties can be measured using a heterodyne reference beam. This is well known as a radar Doppler technique and the principles

are identical. It has thus a great advantage over ordinary intensity-fluctuation spectroscopy (which is also a well known radar technique) in that the spectrum can be asymmetrical, or of arbitrary shape, and is immediately available from the experimental Doppler spectrum. The ordinary intensity-fluctuation spectrum has other advantages, however, which will be discussed in the last section in relation to the measurement of the structure of a turbulent fluid flow.

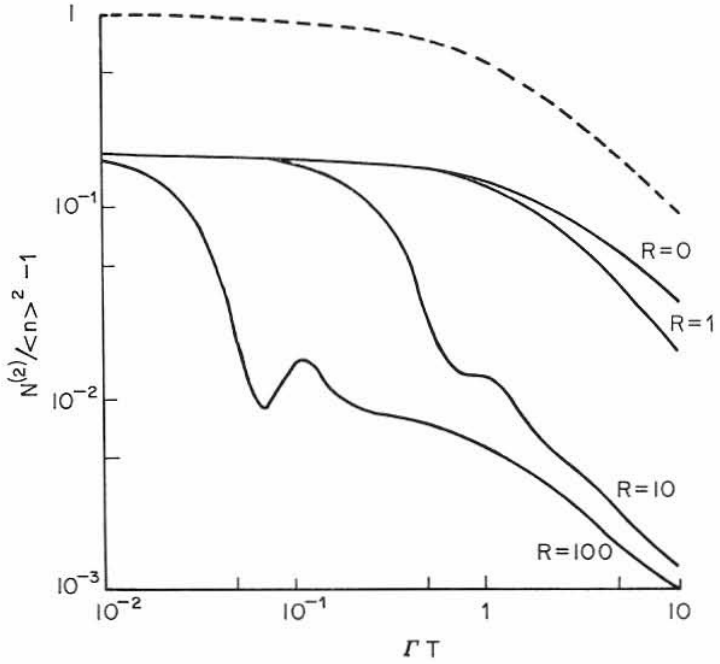


FIG. 14. The excess normalized second factorial moment, $n^{(2)}-1$, $(=N^{(2)}/\langle n\rangle^2-1)$, in the heterodyne case, for the values of $R\,(=|\omega_0-\omega_q|/\Gamma)$ shown and for $\langle n_i\rangle/(\langle n_i\rangle+\langle n_c\rangle)=0{\cdot}1$. The broken lines gives the value for the case where the coherent signal is absent. (After Jakeman and Pike.[58])

4. Photon Statistics of Scattered Laser Light

4.1 Scattering by a random medium

We consider in this section scattering of a coherent laser beam from a randomly inhomogeneous medium. For simplicity we take the medium to be isotropic.

The scattering medium is considered as a scalar field, and the inhomogeneity is described by fluctuations in the local dielectric constant $\varepsilon(\mathbf{r}, t)$. This description is possible because the wavelength of optical radiation is long compared with interatomic spacings. The object of a scattering experiment is to determine the spatial and temporal correlations of this field. These may be written conveniently as

$$C^{(n)}(x_1 \ldots x_{2n}) = \langle \delta\varepsilon^-(x_1) \ldots \delta\varepsilon^-(x_n)\, \delta\varepsilon^+(x_{n+1}) \ldots \delta\varepsilon^+(x_{2n}) \rangle. \tag{66}$$

The question to be asked then, is how are these correlation functions related to the optical field correlations $G^{(n)}$ of light scattered from them? The basic scattering problem was solved by Einstein,[59] Brillouin[60] and Landau and Placzek.[61]

Let us assume an incident field (taking a polarized component),

$$\mathbf{E}_0(\mathbf{r}, t) = \mathscr{E}_0 \exp\,[i(\mathbf{k}_0 \,.\, \mathbf{r} - \omega_0 t)]. \tag{67}$$

We assume the scattering volume is $\gg \lambda^3$, and that the dielectric constant contains fluctuations about a uniform value, ε_0

$$\varepsilon(\mathbf{r}, t) = \varepsilon_0 + \delta\varepsilon(\mathbf{r}, t) \tag{68}$$

The scattered field can then be written

$$\mathbf{E}_s(\mathbf{r}, t) = \mathbf{E}_0(\mathbf{r}, t) + \mathbf{E}_1(\mathbf{r}, t) \tag{69}$$

and Maxwell's equations give in successive orders

$$\nabla^2 \mathbf{E}_0 - \frac{\varepsilon_0}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{E}_0 = 0 \tag{70}$$

$$\nabla^2 \mathbf{E}_1 - \frac{\varepsilon_0}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{E}_1 = \frac{1}{c^2} \frac{\partial^2}{\partial t^2} (\delta\varepsilon\, \mathbf{E}_0) - \frac{\nabla(\nabla \,.\, \delta\varepsilon\, \mathbf{E}_0)}{\varepsilon_0}. \tag{71}$$

The solution at a point $\mathbf{R}$ in the far-field limit is found by a Fourier transform to be

$$E_1(\mathbf{R}_\mathbf{k}, t) = -\frac{{\omega_0}^2}{4\pi c^2} \frac{\mathscr{E}_0 \sin\phi}{R} \exp\,[i\,(\mathbf{k}_s \,.\, \mathbf{R} - \omega_0 t)]\, \delta\varepsilon(\mathbf{K}, t). \tag{72}$$

It is linearly polarized in the plane containing $\mathbf{k}_s$ and $\mathbf{E}_0$. In this formula the direction of the point $\mathbf{R}$ determines the wave vector $\mathbf{k}_s$ of the scattered light and hence also the scattering vector $\mathbf{K}$ which is given by

$$\mathbf{K} = \mathbf{k}_s - \mathbf{k}_0. \tag{73}$$

ϕ is the angle between the polarization direction of $\mathbf{E}_0$ and $\mathbf{R}$. We have therefore,

$$E^+(\mathbf{R}_\mathbf{k}, t) \propto \delta\varepsilon^+(\mathbf{K}, t). \tag{74}$$

Thus a given scattering direction picks out a given spatial Fourier component of the perturbation of the dielectric constant. We form the correlation function

$$\langle E^-(\mathbf{R}_{\mathbf{k}_1}, t_1)\, E^-(\mathbf{R}_{\mathbf{k}_2}, t_2) \ldots E^-(\mathbf{R}_{\mathbf{k}_n}, t_n)\, E^+(\mathbf{R}_{\mathbf{k}_{n+1}}, t_{n+1}) \ldots E^+(\mathbf{R}_{\mathbf{k}_{2n}}, t_{2n})\rangle$$
$$\propto \langle \delta\varepsilon^-(\mathbf{K}_1, t_1)\delta\varepsilon^-(\mathbf{K}_2, t_2) \ldots \delta\varepsilon^-(\mathbf{K}_n, t_n)\delta\varepsilon^+(\mathbf{K}_{n+1}, t_{n+1}) \ldots \delta\varepsilon^+(\mathbf{K}_{2n}, t_{2n})\rangle. \tag{75}$$

This relates the correlation functions G^n of the light field to the correlation functions C^n of the medium (Bertoletti *et al.*).[62] The first-order correlations are of the most direct interest, and in many cases can be calculated by transport theory. From the remarks made in the previous section it should be clear that Doppler spectroscopy offers great advantages here, since when feasible it gives exactly $\langle \delta\varepsilon^-(\mathbf{k}, t_1)\, \delta\varepsilon^+(\mathbf{k}, t_2)\rangle$ for any arbitrary form of this function.

We can see this by the following analysis. The electric field $E(t)$ at the photocathode of the detector in a Doppler (heterodyne) experiment is made up of a reference beam

$$E_r(t) = \mathscr{E}_r \exp(-i\omega_0 t) \tag{76}$$

plus the scattered beam

$$E_s(t) = E_1(\mathbf{K}, t). \tag{77}$$

Thus

$$E(t) = E_r(t) + E_s(t). \tag{78}$$

The measurable output of the photodetector, assuming that the scattered field is statistically stationary, is

$$G^{(2)}(\tau) = \langle I(0)\, I(\tau)\rangle$$
$$= \langle (E_r^-(0) + E_s^-(0))(E_r^-(\tau) + E_s^-(\tau))(E_r^+(\tau) + E_s^+(\tau))(E_r^+(0) + E_s^+(0))\rangle. \tag{79}$$

To simplify this expression we take the case where the complex dielectric constant fluctuations $\delta\varepsilon(\mathbf{K}, t)$ are Gaussian with zero mean. This assumption can be justified in many practical cases.

In these circumstances the expression reduces to

$$G^{(2)}(\tau) = I_r^2 + 2I_r G_s^{(1)}(0) + I_r[G_s^{(1)}(\tau)\exp(-i\omega_0\tau) + c.c.] + G_s^{(2)}(\tau). \tag{80}$$

When passed through a low-frequency reject filter, this signal gives the first-order field correlation function $G_s^{(1)}(\tau)$, from which $C_s^{(1)}(\tau)$ is immediately available

$$C^{(1)}(\tau) \sim \langle \delta\varepsilon^-(\mathbf{K}, \tau)\, \delta\varepsilon^+(\mathbf{K}, 0)\rangle.$$

If the reference beam is removed, the correlation function $G^{(2)}(\tau)$, under the same statistical assumptions, has the same statistical properties as a thermal source and hence we may use the relation given above,

$$G^{(2)}(\tau) = [G^{(1)}(0)]^2 + |G^{(1)}(\tau)|^2, \tag{81}$$

to deduce the first-order correlation from the auto correlation of the photon-counting fluctuations, or the intensity-fluctuation spectrum. I shall give examples of the use of this relation for such studies.

Measurement of higher-order correlations, particularly temporal ones, are not difficult but have had little application as yet. They may possibly prove to be useful for studying scattering in critical phenomena, where Gaussian statistics may not hold.

4.2 APPLICATIONS: INTENSITY-FLUCTUATION AND DOPPLER SPECTROSCOPY

The great forté of the new methods in optical spectroscopy discussed in this chapter is that they allow spectral measurements to be made well beyond the limits of resolution of conventional optical instrumentation. For an optical interference effect to be used, we must have light paths differing by the order of a wavelength; if a resolution of 1 part in 10^{13}, say, is required

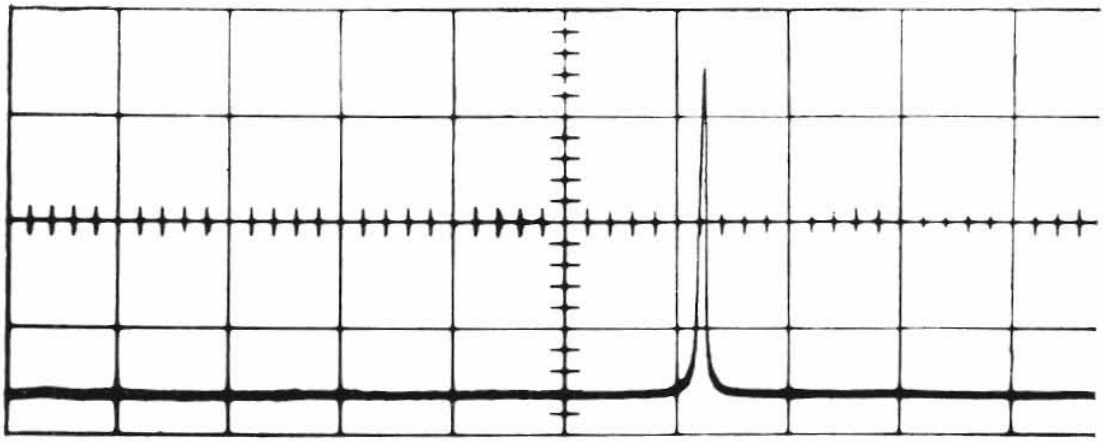


FIG. 15. Scanning Fabry–Pérot oscillogram of a single-mode He–Ne 0·6328 μm laser line. One cm on the screen equals $5{\cdot}0\times10^{-4}$ cm^{-1}. (After Jackson and Pike, *J.Phys. E.*, **1**, 394, 1968).

these paths would be about 10^6m. Even allowing a gain of 100 for multiple reflection such path lengths are not practicable. In Fig. 15 is shown the spectrum of a single-mode laser obtained at R.R.E. with a 1m long Fabry–Pérot interferometer working at a finesse of 150. The line width is instrumentally limited at 1 MHz and represents an optical resolution of only ~ 2 in 10^9. With scattered radiation even 200 MHz resolution is difficult to achieve. In Fig. 16 the Rayleigh and Brillouin lines of benzene are shown at approximately this resolution (see also Jackson *et al.*[63]). We see that if phenomena taking place on a time scale less than megacycles per second are to be studied by light scattering, using the intrinsically narrow (few Hz) lines from laser sources, then the techniques of statistical optics must be used.

Strictly we should not include Doppler spectroscopy as a statistical technique since, although the intensity fluctuation of the mixed field is measured, the results are independent of the statistical properties of the source. We include it, however, as an example of the use of intensity-fluctuations. We

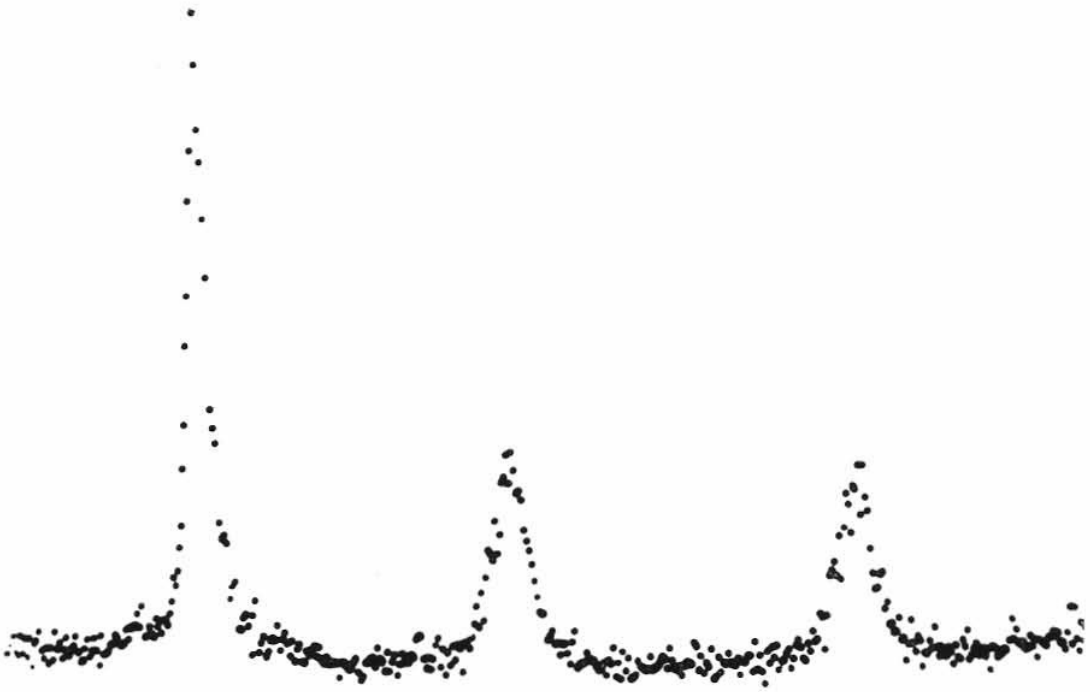


FIG. 16. Brillouin and Rayleigh spectrum of benzene by Fabry–Pérot etalon with digital data storage, showing the decay times of acoustic photons and of entropy fluctuations.

should now ask, conversely, what are the limits of intensity-fluctuation spectroscopy? Here one is limited solely by the response time of existing detectors. The fastest photomultiplier tubes available have pulse rise times something less than one nanosecond. Beat frequencies greater than a GHz will thus be averaged out. Thus there is a range of phenomena, having time dependencies between, say, 1 MHz and 1 GHz, which can be studied by either technique. We have already seen above, in the results of experiments on laser statistics, the study of fluctuations in the microsecond region by statistical spectroscopy. We have already mentioned the work of Forrester *et al.*,[1] which was in the microwave region. The mercury discharge lamp has been the subject of study by statistical methods by a number of workers (Hanbury-Brown and Twiss,[2] Rebka and Pound,[64] Brannen *et al.*,[65] Twiss and Little,[66] Johnson *et al.*,[20] Morgan and Mandel[67]). The width

of such a line ($\sim$200 MHz) lies at the high end of the frequency scale for statistical techniques and it is usually better to make such measurements using optical methods. The results of the last-quoted paper obtained by single-channel coincidence are shown in Fig. 17. In the penultimate reference

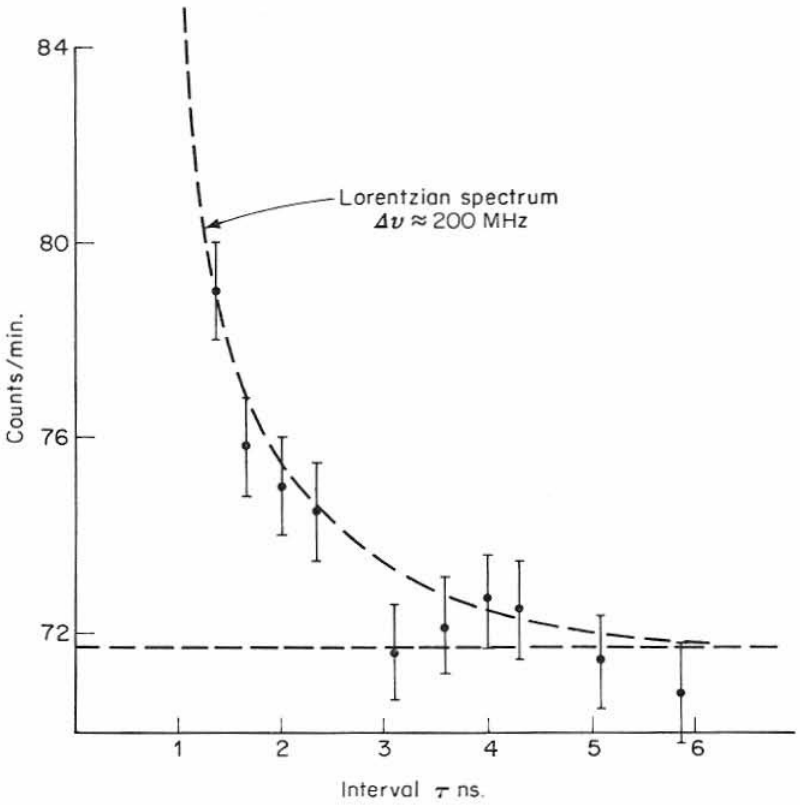


FIG. 17. Mercury arc lamp photon coincidences. (After Morgan and Mandel.[67])

the measurement was made by the photon-counting distribution. Measurements become progressively easier but less feasible by optical means as we come down in frequency. Before leaving the applications of these techniques to natural sources, however, I should remind you of the most famous and perhaps most important application in this category, which is the Narrabri stellar interferometer (Hanbury-Brown[68]). This is now well in operation and can measure stellar diameters down to about 5×10^{-4} second of arc.

In Fig. 18 is given an example of laser Doppler spectroscopy at 60 MHz. This is Brillouin scattering from an acousto-electric oscillator (phonon-maser). This Doppler laser-scattering application[77] has allowed a measurement of the soundwave flux inside the oscillator to be obtained, which is extremely difficult to achieve by any other method.

From Professor Cummins' laboratory at John Hopkins University, I give an example[69] of the use of intensity-fluctuation spectroscopy in the study of transport parameters of CO_2 near its critical point. Typical results

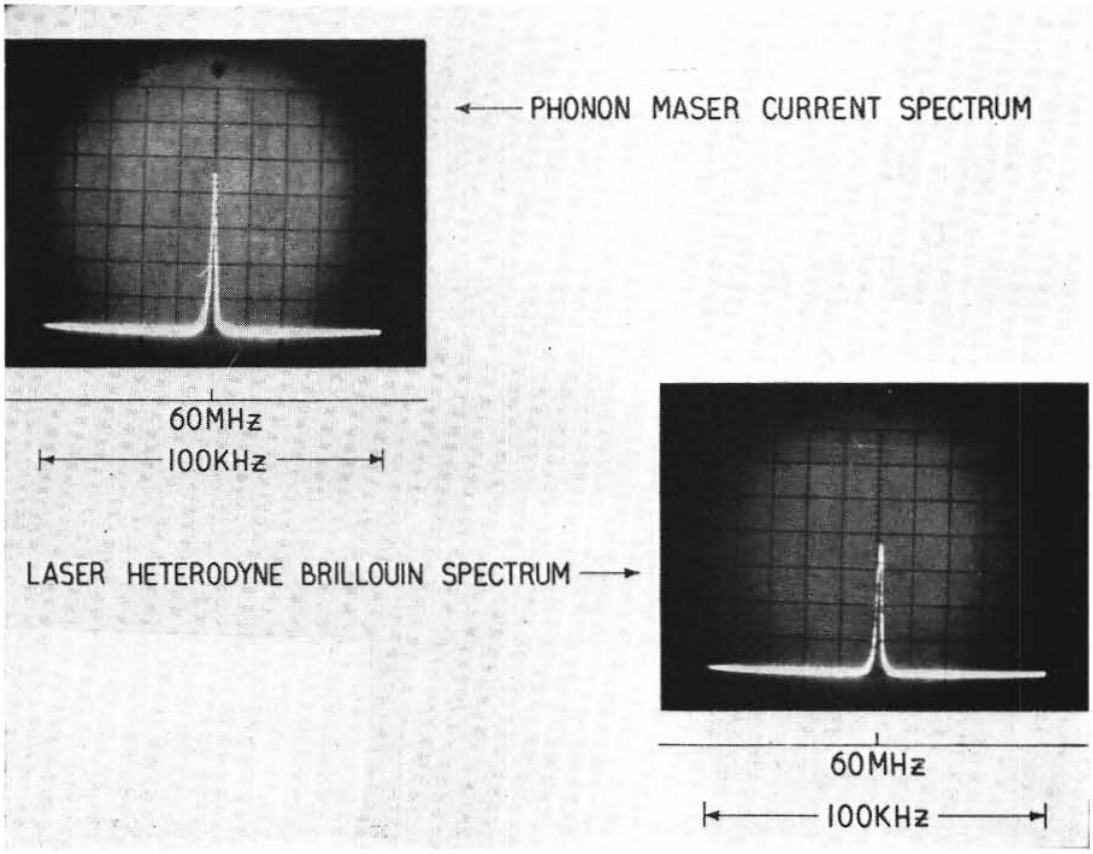


FIG. 18. The Brillouin spectrum of an acousto-electric oscillator by Doppler spectroscopy.

are shown in Fig. 19. This very beautiful work and similar work in Professor Benedek's laboratory at M.I.T., particularly by Saxman,[70] has provided a great experimental move forward in the subject of the study of critical phenomena. The method used in general is to analyse the intensity-fluctuation spectrum of the Rayleigh scattered light using a wave analyser, although Cummins[71] has used an analogue correlator. I should say that a wealth of original experimental work in this new area of spectroscopy has come from these two laboratories.

Another example where statistical spectroscopy has made an impact on a subject where optics previously was of limited application, has been in the study of turbulence (Goldstein *et al.*,[72] Pike *et al.*,[73] Bourke *et al.*[74]).

In Fig. 20 is shown an experimental arrangement for the study of the structure of turbulent flow of a fluid by intensity-fluctuation spectroscopy. The spectrum of the scattered light contains "Doppler-shifted" frequencies corresponding to the different velocities present in a "coherence volume".

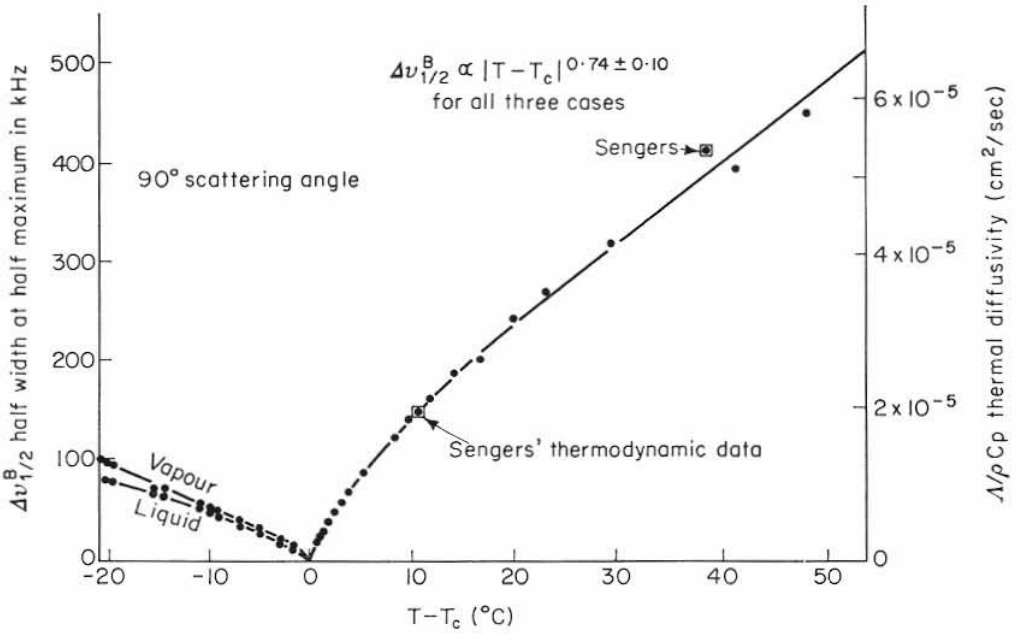


FIG. 19. Rayleigh linewidth along critical isochore for CO_2 by intensity-fluctuation spectroscopy. (After Swinney and Cummins, reproduced by permission of the American Physical Society from their Bulletin.)

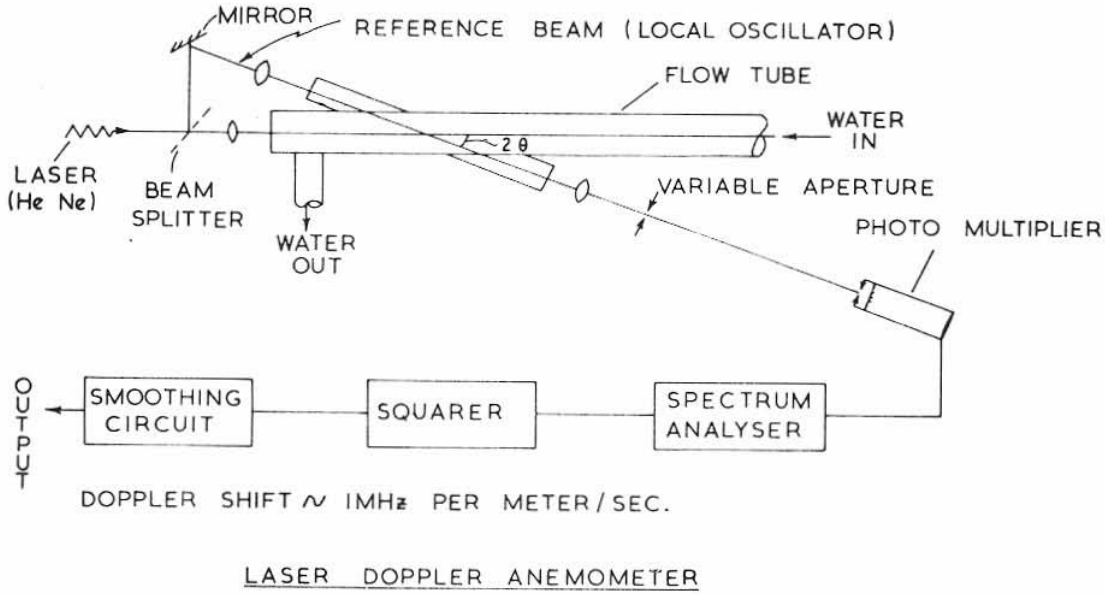


FIG. 20. Apparatus for studying the structure of turbulent flow by intensity-fluctuation spectroscopy.

This volume is controlled by geometrical factors and can be varied from small to large compared with the scale of the turbulence. Results obtained recently at R.R.E. are shown in Figs. 21 and 22. Interpretation of these shows that the velocity distribution is closely Gaussian; its width in the large-volume case agrees with previous determinations by hot-wire anemometry. For intermediate volumes, such work has shown that the scale of turbulence can be quantitatively measured. The method used was again spectrum analysis by wave analyser.

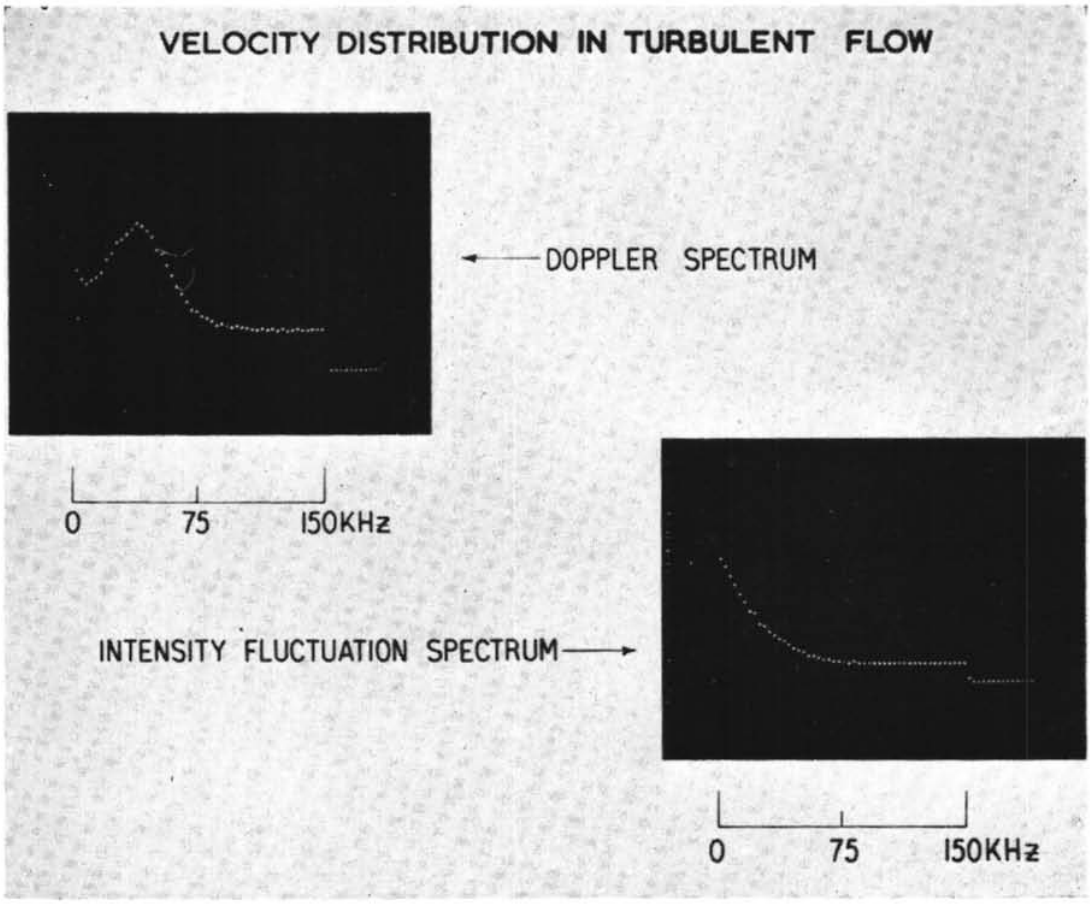


FIG. 21. Doppler and intensity-fluctuation spectra of turbulent flow.

My last example[75] gives another illustration of the power of the method of single clipped autocorrelation of photon-counting fluctuations, known locally as SCAC. It is here applied to the determination of translational diffusion coefficients, D_T, of protein molecules in aqueous solution, a subject which has been pioneered in the laboratories of Professors Cummins and Benedek. The apparatus is shown in Fig. 23. The diffusion constant controls

the Brownian motion which can be measured by determining the optical spectrum of scattered laser light. The theoretical prediction for a protein

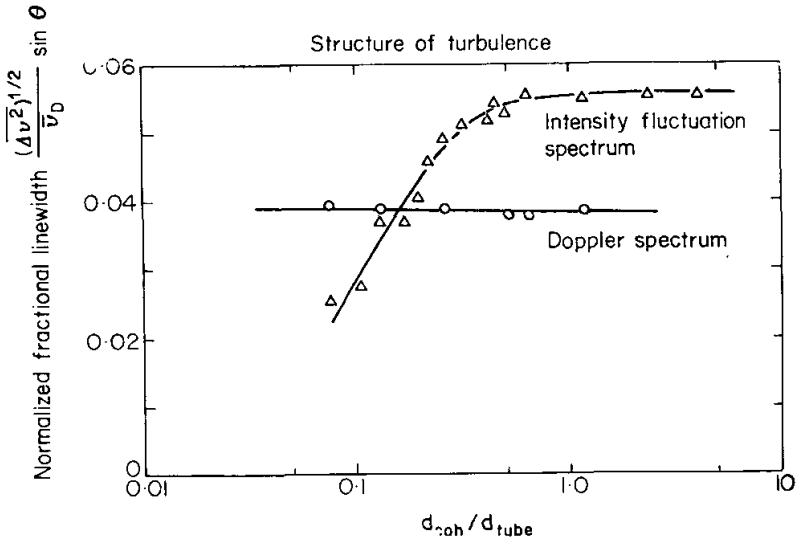


FIG. 22. Linewidths of intensity-fluctuation and Doppler spectra of turbulent flow as a function of coherence volume. (After Bourke *et al.*[74])

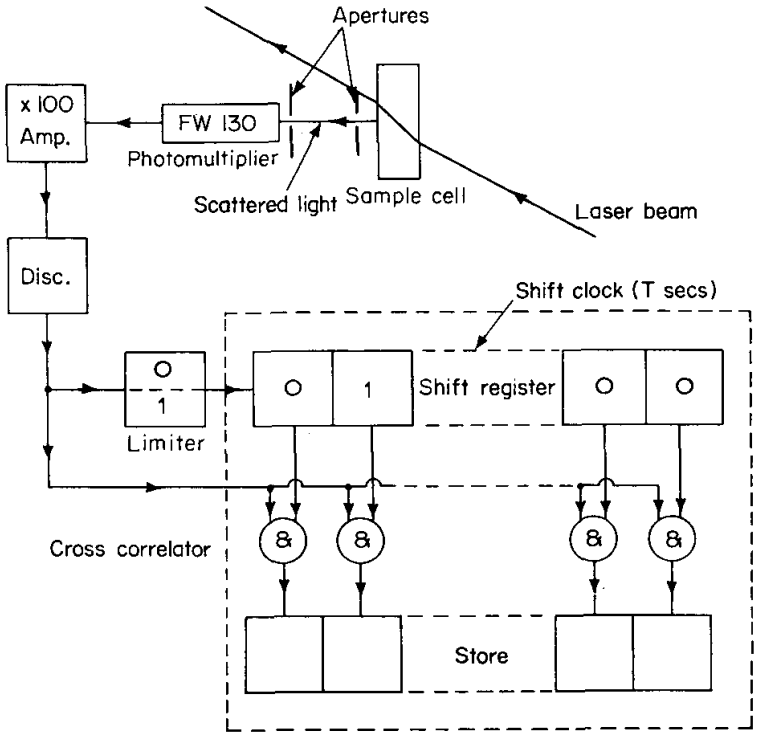


FIG. 23. Apparatus for measuring the single-clipped autocorrelation of photon-counting fluctuations. (After Foord *et al.*[75])

small compared with the optical wavelength is a single exponential with decay time

$$\tau_c = \tfrac{1}{2} D_T |\mathbf{K}|^2, \tag{82}$$

where $\mathbf{K}$ is the scattering vector.

A typical result of a SCAC experiment is given in Fig. 24 for the protein bovine serum albumin. About 40 mW of He–Ne multimode laser light was focussed in the sample cell where a concentration of 1 mg/ml of the protein was suspended in a universal buffer solution of controlled pH. Light was collected at a scattering angle of approximately 30° from about 10% of a coherence area. The total photoelectron counting rate was 200 counts per

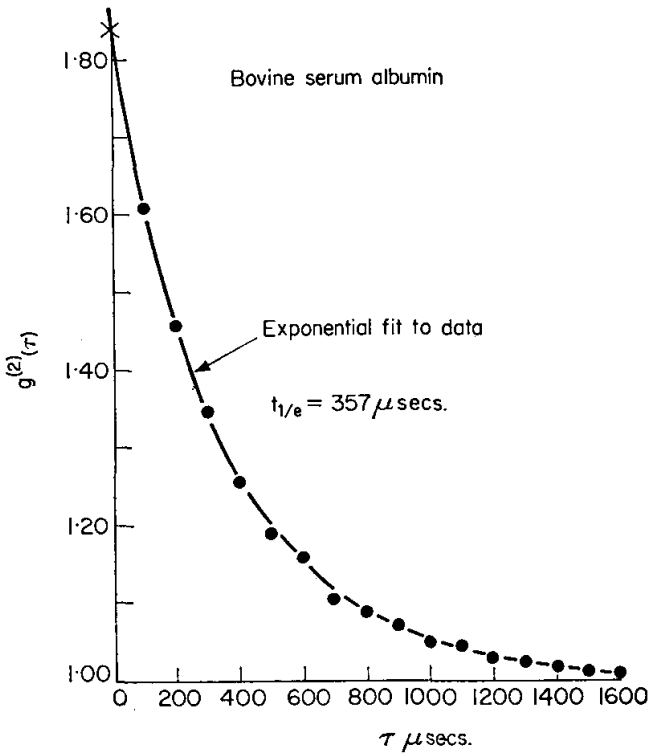


FIG. 24. The second-order correlation function, $g^{(2)}(\tau)$ of laser light scattered from the protein bovine serum albumin obtained by the method of single-clipped autocorrelation of photon counting fluctuations. (After Foord *et al.*[75])

second. An accuracy of 2% can be obtained in an experiment lasting one hour. The important difference made possible by the SCAC method over the wave analyser is that for a given experimental time, the concentration of the protein can be reduced in the former case by the ratio of the bandwidth of the wave analyser to the frequency range scanned. Nearly two orders of magnitude can be gained in this way. Since the diffusion constant

is dependent on concentration, this is a very important consideration. In Fig. 25 are shown some preliminary results we have obtained by varying the pH of the buffer solution. The protein can be seen to "uncurl", that is, its diffusion coefficient increases as the pH is reduced. Experiments along these

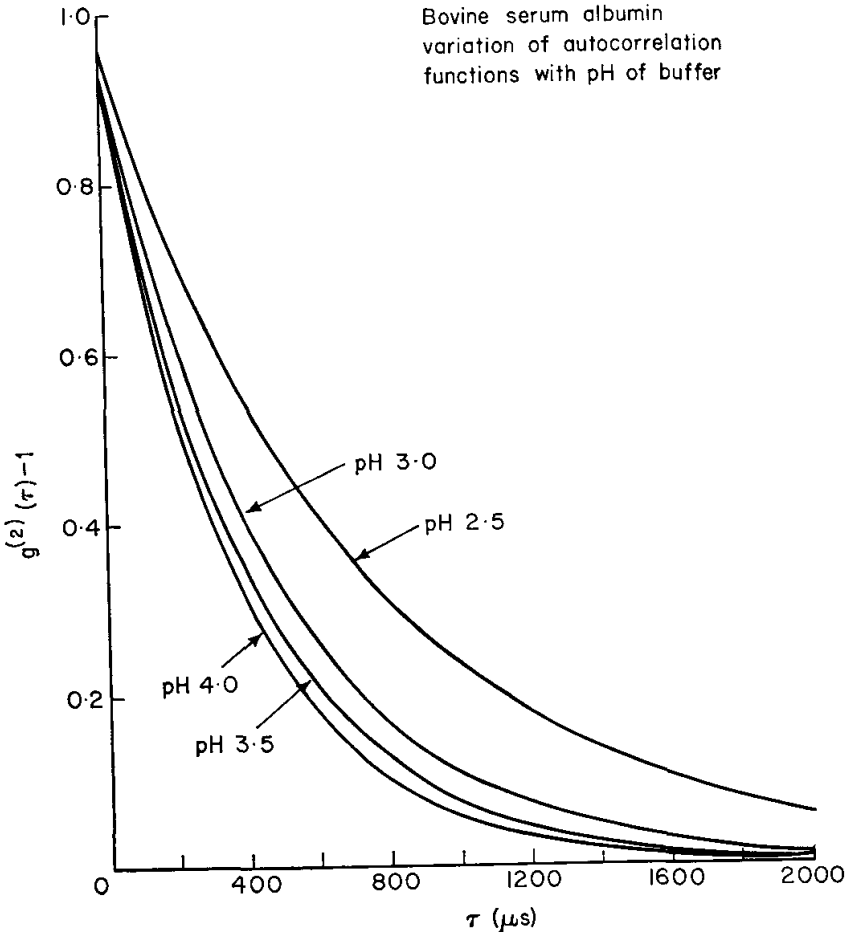


FIG. 25. The effect of changing pH on the diffusion constant (and hence size) of the protein bovine serum albumin using the apparatus of Fig. 23.

lines have aroused the interest of a number of biologists; the relative speed and accuracy of the measurements open up new possibilities in the study of time-dependent processes which will no doubt prove rewarding in the future.

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Quantum-Classical Correspondence for Stochastic Processes

W. H. LOUISELL

The statistical properties of the radiation field in a laser and optical parametric oscillator have been important in the understanding of these devices. Recently, the techniques developed to study the laser have been applied with gratifying success to the study of the noise properties of Josephson junction oscillators. Some of these methods are presented in this chapter. It is believed they will prove very useful in the future, in other areas of physics.

Section 1 contains a brief review of several topics in quantum mechanics.[1] In Section 2 the quantum theory of damping under the Markoff approximation is presented. The concept of a reduced density operator is introduced which describes the statistical properties of a system coupled to a damping mechanism. This is applied to a damped mode of the radiation field.

The concept of ordered operators as a means of developing a quantum-classical correspondence is presented in Section 3. This allows us to treat quantum stochastic processes in an equivalent classical domain.[2] A quantum regression theorem due to Lax is also derived which allows us to convert multi-time averages to one-time averages for Markoffian systems.[2,3] The spectra of the radiation field can then be determined.

In the final section these techniques are applied to a rotating wave van der Pol oscillator. A single mode homogeneously broadened laser has been shown to be equivalent to such an oscillator.[3]

1. Review of Quantum Mechanics

1.1. Equations of Motion

a. *The Schrödinger Equation*

The Schrödinger equation in Dirac notation is

$$i\hbar \frac{\partial |\psi(t)\rangle}{\partial t} = H|\psi(t)\rangle, \tag{1}$$

where the Hamiltonian is Hermitian

$$H = H^{\dagger}. \tag{2}$$

When H is time independent, the solution of (1) is

$$|\psi(t)\rangle = \exp(-iHt/\hbar)\,|\psi(0)\rangle. \tag{3}$$

This state vector is normalized to unity so that

$$\langle\psi(t)\,|\,\psi(t)\rangle = 1. \tag{4}$$

b. *Heisenberg Equation of Motion*

If p_ψ is the probability of the system being in state ψ and M is any system operator, the mean value of M is defined by

$$\begin{aligned}\langle M(t)\rangle &= \sum_\psi p_\psi \langle\psi(t)\,|\,M\,|\,\psi(t)\rangle \\ &= \sum_\psi p_\psi \langle\psi(0)\,|\exp(iHt/\hbar)\,M\exp(-iHt/\hbar)\,|\,\psi(0)\rangle,\end{aligned} \tag{5}$$

where

$$\sum_\psi p_\psi = 1. \tag{6}$$

Here, $\langle\psi(t)\,|\,M\,|\,\psi(t)\rangle$ is a quantum ensemble average when the system is known to be in state ψ, while the second average over states ψ is due to the classical like uncertainty in the state ψ. If we define the Heisenberg operator as

$$M_H(t) = \exp(iHt/\hbar)\,M\exp(-iHt/\hbar) \tag{7}$$

then

$$\langle M(t)\rangle = \sum_\psi p_\psi \langle\psi(0)\,|\,M_H(t)\,|\,\psi(0)\rangle. \tag{8}$$

From (7) we see that

$$\begin{aligned}i\hbar\frac{\mathrm{d}M_H(t)}{\mathrm{d}t} &= \exp(iHt/\hbar)\,M\exp(-iHt/\hbar)\,H - H\exp(iHt/\hbar)\,M\exp(-iHt/\hbar) \\ &= [M_H(t), H]\end{aligned} \tag{9}$$

which is the Heisenberg equation of motion for the operator in the Heisenberg picture.

c. *Density Operator Equation of Motion*

We have the relation

$$\mathrm{Tr}\,|u\rangle\langle v| = \langle v\,|\,u\rangle. \tag{10}$$

To prove this we may use any complete set of states such that

$$\sum_n |n\rangle\langle n| = 1. \tag{11}$$

Then

$$\mathrm{Tr}\,|u\rangle\langle v| = \sum_n \langle n\,|\,u\rangle\langle v\,|\,n\rangle = \sum_n \langle v\,|\,n\rangle\langle n\,|\,u\rangle = \langle v\,|\,u\rangle.$$

This result allows us to re-write (5) as

$$\langle M(t)\rangle = \mathrm{Tr}\left\{M\sum_{\psi} p_\psi\,|\psi(t)\rangle\langle\psi(t)|\right\}. \tag{12}$$

We then define the density operator $\rho(t)$ in the Schrödinger picture by

$$\rho(t) = \sum_{\psi} p_\psi\,|\psi(t)\rangle\langle\psi(t)|, \tag{13}$$

so that

$$\langle M(t)\rangle = \mathrm{Tr}\,M\rho(t). \tag{14}$$

M and $\rho(t)$ are both in the Schrödinger picture.

If we use (1) and its adjoint, it follows from (13) that

$$\begin{aligned} i\hbar\frac{\partial\rho}{\partial t} &= \sum_{\psi} p_\psi\left\{i\hbar\frac{\partial|\psi(t)\rangle}{\partial t}\langle\psi(t)| + i\hbar\,|\psi(t)\rangle\,\frac{\partial}{\partial t}\langle\psi(t)|\right\} \\ &= \sum_{\psi} p_\psi\,\{H\,|\psi(t)\rangle\langle\psi(t)| - |\psi(t)\rangle\langle\psi(t)|\,H\} \\ &= [H,\rho(t)] \end{aligned} \tag{15}$$

which is the equation obeyed by the density operator. Again if H is time independent, the solution is

$$\rho(t) = \exp(-iHt/\hbar)\,\rho(0)\exp(iHt/\hbar) = \sum_{\psi} p_\psi\,|\psi(t)\rangle\langle\psi(t)|. \tag{16}$$

Therefore, by (3) and its adjoint we see that

$$\rho(0) = \sum_{\psi} p_\psi\,|\psi(0)\rangle\langle\psi(0)|. \tag{17}$$

Furthermore, by (10), (6) and (4) we see that

$$\mathrm{Tr}\,\rho(t) = \sum_{\psi} p_\psi\,\langle\psi(t)\,|\,\psi(t)\rangle = \sum_{\psi} p_\psi = 1. \tag{18}$$

We note next that

$$\mathrm{Tr}\,AB = \mathrm{Tr}\,BA. \tag{19}$$

This follows if we use (11):

$$\begin{aligned} \mathrm{Tr}\,AB &= \sum_{n}\langle n\,|\,AB\,|\,n\rangle = \sum_{n,m}\langle n\,|\,A\,|\,m\rangle\,\langle m\,|\,B\,|\,n\rangle \\ &= \sum_{n,m}\langle m\,|\,B\,|\,n\rangle\langle n\,|\,A\,|\,m\rangle = \sum_{m}\langle m\,|\,BA\,|\,m\rangle \\ &= \mathrm{Tr}\,BA \end{aligned}$$

By (14), (16) and (19), we see that

$$\langle M(t)\rangle = \mathrm{Tr}\, M\, \rho(t) = \mathrm{Tr}\, M \exp(-iHt/\hbar)\, \rho(0) \exp(iHt/\hbar)$$
$$= \mathrm{Tr} \exp(iHt/\hbar)\, M \exp(-iHt/\hbar)\, \rho(0) = \mathrm{Tr}\, M_H(t)\, \rho(0). \tag{20}$$

Therefore, traces may be evaluated in the Schrödinger or Heisenberg pictures.

1.2. THE RADIATION FIELD

a. *Quantization*

The radiation field without sources may be described classically in mks units by the vector potential $\mathbf{A}(\mathbf{r}, t)$ which obeys the wave equation

$$\nabla^2 \mathbf{A}(\mathbf{r}, t) = \frac{1}{c^2} \frac{\partial^2 \mathbf{A}}{\partial t^2}(\mathbf{r}, t). \tag{21}$$

The electric and magnetic fields are

$$\mathbf{E}(\mathbf{r}, t) = \frac{-\partial \mathbf{A}}{\partial t} \tag{22}$$

$$\mathbf{B}(\mathbf{r}, t) = \mathrm{curl}\, \mathbf{A}.$$

We may expand $\mathbf{A}$ in the form

$$\mathbf{A}(\mathbf{r}, t) = \frac{1}{\sqrt{\varepsilon_0}} \sum_l q_l(t)\, \mathbf{u}_l(\mathbf{r}), \tag{23}$$

where

$$\frac{\mathrm{d}^2 q_l}{\mathrm{d}t^2} + \omega_l^2\, q_l = 0 \tag{24}$$

$$\nabla^2 \mathbf{u}_l(\mathbf{r}) + \left(\frac{\omega_l}{c}\right)^2 \mathbf{u}_l(\mathbf{r}) = 0. \tag{25}$$

When we solve (25) in a cavity with perfectly conducting walls, we obtain a set of normal modes. The $q_l(t)$ give the amplitude of the lth mode and obey the equation of motion of a simple harmonic oscillator. If we define

$$p_l(t) = \frac{\mathrm{d}q_l}{\mathrm{d}t} \tag{26}$$

then the electric and magnetic fields become

$$\mathbf{E}(\mathbf{r}, t) = \frac{-1}{\sqrt{\varepsilon_0}} \sum_l p_l(t)\, \mathbf{u}_l(\mathbf{r}). \tag{27}$$

$$\mathbf{B}(\mathbf{r}, t) = \frac{1}{\sqrt{\varepsilon_0}} \sum_l q_l(t) \operatorname{curl} \mathbf{u}_l(\mathbf{r}). \tag{28}$$

so that $p_l(t)$ determines the electric field and $q_l(t)$ the magnetic field.

The normal modes of (25) are orthogonal and normalized to unity:

$$\int_{\text{cavity}} \mathbf{u}_l^*(\mathbf{r}) \cdot \mathbf{u}_m(\mathbf{r})\, d\mathbf{r} = \delta_{lm}. \tag{29}$$

The energy of the field in the cavity is

$$\begin{aligned} H &= \int_{\text{cavity}} \left[\frac{\varepsilon_0}{2} \mathbf{E}^2(\mathbf{r}, t) + \frac{1}{2\mu_0} \mathbf{B}^2(\mathbf{r}, t)\right] d\mathbf{r} \\ &= \sum_l \tfrac{1}{2}(p_l^2 + \omega_l^2 q_l^2), \end{aligned} \tag{30}$$

which follows from (27–29). We may therefore visualize the electromagnetic field energy as residing in an infinite set of fictitious harmonic oscillators, one for each mode.

To quantize the field, we merely quantize the oscillators. We assume p_l and q_l are Hermitian operators which obey the commutation relations,

$$[q_l, p_m] = i\hbar \delta_{lm}. \tag{31}$$

If we introduce the linear combinations

$$a = \frac{1}{\sqrt{(2\hbar\omega)}} (\omega q + ip) \qquad q = \sqrt{\left(\frac{\hbar}{2\omega}\right)} (a^\dagger + a)$$

or (32)

$$a^\dagger = \frac{1}{\sqrt{(2\hbar\omega)}} (\omega q - ip) \qquad p = i\sqrt{\left(\frac{\hbar\omega}{2}\right)} (a^\dagger - a)$$

for each mode, it follows from (31) that

$$[a_l, a_m^\dagger] = \delta_{lm}. \tag{33}$$

The Hamiltonian operator (30) then reduces to

$$H = \sum_l \hbar\omega_l (a_l^\dagger a_l + \tfrac{1}{2}). \tag{34}$$

b. *Eigenvalues and Eigenvectors.*

If we restrict ourselves for simplicity to one mode, the Schrödinger equation (1) becomes

$$i\hbar \frac{\partial}{\partial t} |\psi(t)\rangle = \hbar\omega(a^\dagger a + \tfrac{1}{2}) |\psi(t)\rangle. \tag{35}$$

Since the Hamiltonian is time independent, we may separate variables. Thus, if we let

$$|\psi(t)\rangle = \exp(iEt/\hbar) |\psi(0)\rangle, \tag{36}$$

we obtain the eigenvalue equation

$$\hbar\omega(a^\dagger a + \tfrac{1}{2}) |\psi(0)\rangle = E |\psi(0)\rangle, \tag{37}$$

where E is the energy eigenvalue.

Since $(a^\dagger a)^\dagger = a^\dagger a$, this operator is Hermitian and has real eigenvalues. We may write this as

$$a^\dagger a |n'\rangle = n' |n'\rangle, \tag{38}$$

where we label the eigenvector by the corresponding eigenvalue.

To solve, we note by (33) that

$$[a, a^\dagger a] = a \qquad [a^\dagger, a^\dagger a] = -a^\dagger \tag{39a}$$

or

$$a^\dagger a a = a(a^\dagger a - 1); \qquad a^\dagger a a^\dagger = a^\dagger(a^\dagger a + 1). \tag{39b}$$

If we use these to operate on $|n'\rangle$, we have by (38) that

$$a^\dagger a\{a |n'\rangle\} = (n'-1)\{a |n'\rangle\}, \tag{40}$$

$$a^\dagger a\{a^\dagger |n'\rangle\} = (n'+1)\{a^\dagger |n'\rangle\}. \tag{41}$$

Therefore, if $|n'\rangle$ is an eigenvector of $a^\dagger a$ with eigenvalue n', then $a|n'\rangle$ is also an eigenvector of $a^\dagger a$ with eigenvalue $n'-1$ while $a^\dagger |n'\rangle$ is an eigenvector with eigenvalue $n'+1$. We may therefore proceed to generate an infinite set of eigenvectors and eigenvalues from the assumed original one:

$$\begin{cases} |n'\rangle & a|n'\rangle & a^2|n'\rangle & \dots \\ n' & n'-1 & n'-2 & \dots \end{cases}$$

$$\begin{cases} |n'\rangle & a^\dagger|n'\rangle & a^{\dagger 2}|n'\rangle & \dots \\ n' & n'+1 & n'+2 & \dots \end{cases}$$

We next show that $n' = 0, 1, 2, \dots$. The length of a vector must be positive. Therefore,

$$\langle n' | n' \rangle \geqslant 0.$$

If the equality holds, then $|n'\rangle = 0$ which is uninteresting and trivial. Therefore, $\langle n' | n' \rangle > 0$. Next, the length of $a|n'\rangle$ is

$$\langle n' | a^\dagger a | n' \rangle = n' \langle n' | n' \rangle \geqslant 0,$$

where we again used (38) and the fact that $(a|n'\rangle)^\dagger = \langle n'|a^\dagger$. Since $\langle n'|n'\rangle > 0$, then $n' \geqslant 0$. Therefore, all the real eigenvalues n' must be non-negative. The generated sequence $n', n'-1, n'-2, \ldots$ must be terminated before reaching negative values. Therefore, n' must take values 0, 1, 2, ...

To normalize the vectors, we write

$$a|n\rangle = c_n|n-1\rangle.$$

Then

$$\langle n|a^\dagger a|n\rangle = |c_n|^2 \langle n-1|n-1\rangle = n\langle n|n\rangle.$$

If we require that

$$\langle n|n\rangle = 1$$

$$\langle n-1|n-1\rangle = 1,$$

then aside from a trivial phase, $c_n = \sqrt{n}$ and we have

$$a|n\rangle = \sqrt{n}|n-1\rangle \qquad (n = 0, 1, 2, \ldots). \tag{42}$$

In a similar way, we see that

$$a^\dagger|n\rangle = \sqrt{(n+1)}|n+1\rangle \tag{43}$$

and

$$a^\dagger a|n\rangle = n|n\rangle \tag{44}$$

and the energy eigenvalues are

$$E_n = \hbar\omega(n+\tfrac{1}{2}). \tag{45}$$

It follows from (43) by repeated application of $a^\dagger$ to the vacuum or ground state $|0\rangle$ that

$$\frac{a^{\dagger l}}{\sqrt{l!}}|0\rangle = |l\rangle. \tag{46}$$

If the field is in the energy eigenstate $|n\rangle$ with energy $E_n = \hbar\omega(n+\frac{1}{2})$, we may say that the cavity has n units of excitation $\hbar\omega$. Each unit of excitation may be called a quantum or photon and then one says there are exactly n photons in the cavity of energy $\hbar\omega$. When a operates on $|n\rangle$, we have the state $|n-1\rangle$ with one less photon. We say that a "annihilates" a photon. Similarly, since $a^\dagger|n\rangle = \sqrt{(n+1)}|n+1\rangle$, $a^\dagger$ "creates" a photon.

It is not difficult to show that the photon eigenstates are orthogonal

$$\langle n|m\rangle = \delta_{mn} \tag{47}$$

and complete

$$\sum_{n=0}^{\infty} |n\rangle\langle n| = 1 \tag{48}$$

where 1 is the identity operator.

c. *Heisenberg Equations of Motion*

The Heisenberg equation of motion for $a(t)$ is

$$i\hbar \frac{da}{dt} = [a, H] = \hbar\omega[a, a^\dagger a] = \hbar\omega a, \tag{49}$$

where we used (34) and (39a). The solution is

$$a(t) = \exp(-i\omega t)\, a(0) = \exp(i\omega t a^\dagger a)\, a \exp(-i\omega t a^\dagger a), \tag{50}$$

where the last form follows from (7). Similarly, one finds that

$$a^\dagger(t) = \exp(i\omega t)\, a^\dagger(0) = \exp(i\omega t a^\dagger a)\, a^\dagger \exp(-i\omega t a^\dagger a). \tag{51}$$

2. Quantum Theory of Damping

2.1. The reduced density operator under Markoff approximation

a. *Arbitrary System*

Consider a system described by a Hamiltonian, H, a reservoir described by a Hamiltonian, R, and a weak interaction between the system and reservoir V. The density operator for the system and reservoir, $\rho(t)$, in the Schrödinger picture satisfies the equation

$$\frac{\partial \rho}{\partial t} = \frac{1}{i\hbar}[H + R + V, \rho]. \tag{52}$$

In general, we are interested in mean values of a function, M, of the system operators only. This is given in the Schrödinger picture by

$$\langle M(t)\rangle = \mathrm{Tr}_{R,S}\, M\, \rho(t) = \mathrm{Tr}_S[M\, \mathrm{Tr}_R\, \rho(t)], \tag{53}$$

where we have traced over both the system and reservoir. From this we conclude that we never need all the information contained in $\rho(t)$, but that we need only the reduced density operator for the system defined by

$$S(t) = \mathrm{Tr}_R\, \rho(t), \tag{54}$$

where we trace $\rho(t)$ over the reservoir. $S(t)$ is an operator function of system operators only. We would therefore like to remove the unnecessary information in (52) and obtain an equation of motion for $S(t)$ directly.

To proceed, it is most convenient to transform (52) into the interaction

picture. Physically, this removes the high frequency unperturbed motion from the equation of motion. To this end we let

$$\rho(t) = \exp\left[\frac{-i}{\hbar}(H+R)t\right]\chi(t)\exp\left[\frac{i}{\hbar}(H+R)t\right]. \tag{55}$$

This represents a transformation from the Schrödinger picture, $\rho(t)$, to the interaction picture, $\chi(t)$. Since the system and reservoir are independent before coupling, we note that $[H, R] = 0$. In fact, all system operators commute with all reservoir operators.

If we substitute (55) into (52), we find that $\chi(t)$ satisfies the equation

$$i\hbar\frac{\partial\chi}{\partial t} = [V(t), \chi] \tag{56}$$

where

$$V(t) = \exp\left[\frac{i}{\hbar}(H+R)t\right]V\exp\left[\frac{-i}{\hbar}(H+R)t\right]. \tag{57}$$

If we trace both sides of (55) over the reservoir and use (54) we have

$$\begin{aligned} S(t) &= \mathrm{Tr}_R \exp\left[\frac{-i}{\hbar}(H+R)t\right]\chi(t)\exp\left[\frac{i}{\hbar}(H+R)t\right] \\ &= \exp\left(\frac{-i}{\hbar}Ht\right)s(t)\exp\left(\frac{i}{\hbar}Ht\right), \end{aligned} \tag{58}$$

where

$$s(t) = \mathrm{Tr}_R\,\chi(t) \tag{59}$$

is the reduced system density operator in the interaction picture.

We assume that the interaction is turned on at $t = 0$. At that time, the system and reservoir are independent, so that the density operator factorizes into the direct product

$$\rho(0) = S(0)f_0(R) = s(0)f_0(R) = \chi(0), \tag{60}$$

where $S(0)$ is the initial system density operator and $f_0(R)$ is the initial reservoir density operator. The reservoir is assumed to be in thermal equilibrium at temperature T so that

$$f_0(R) = \frac{\exp(-\beta R)}{\mathrm{Tr}_R \exp(-\beta R)}, \tag{61}$$

where $\beta = (kT)^{-1}$, a Boltzmann distribution. In addition, we assume that the reservoir is so large that its statistical properties are unaffected by the weak coupling to the system. In other words, we assume the reservoir remains in

equilibrium when it is coupled to the system. The assumption that the interaction is "turned on" at $t = 0$ is just an artifice to remove all statistical correlations between the system and reservoir initially.

We return to (56) and integrate both sides from $t = 0$ to t subject to the initial value (60). This gives on two iterations

$$\chi(t) = \chi(0) + \frac{1}{i\hbar}\int_0^t [V(t'), \chi(0)]\,dt' + \left(\frac{1}{i\hbar}\right)^2 \int_0^t dt' \int_0^t dt'' \left[V(t'), [V(t''), \chi(t'')]\right]. \tag{62}$$

If we proceeded to iterate in this way, we would obtain $\chi(t)$ in a power series in the perturbation $V(t)$ which is just a perturbation result. Unfortunately, perturbation theory is not adequate to yield exponential decay and we resort to another approximation. If we differentiate both sides of (62) with respect to t, we have

$$\frac{\partial \chi}{\partial t} = \frac{1}{i\hbar}[V(t), \chi(0)] + \left(\frac{1}{i\hbar}\right)^2 \int_0^t \left[V(t), [V(t''), \chi(t'')]\right] dt''. \tag{63}$$

If we trace both sides over the reservoir and use (59) we obtain

$$\frac{\partial s}{\partial t} = \left(\frac{1}{i\hbar}\right)^2 \int_0^t \mathrm{Tr}_R \left[V(t), [V(t'), \chi(t')]\right] dt', \tag{64}$$

where we have assumed that

$$\mathrm{Tr}_R\, V(t)\,\chi(0) = \mathrm{Tr}_R\, \chi(0)\, V(t) = 0$$

and $\chi(0) = S(0) f_0(R)$. That is, we assume that the interaction has no diagonal elements in the representation in which R is diagonal. If V does have diagonal terms, we could re-define the system Hamiltonian to include them, so this is no real specialization.

At this point we assume that $V \ll H$ or R. If $V = 0$, the system and reservoir are independent and the density operator $\chi(t)$ would factorize into the direct product $\chi(t) = s f_0$ where we assume the reservoir remains in equilibrium. Since V is small, we therefore look for a solution of (64) of the form

$$\chi(t) = s(t) f_0(R) + \Delta\chi(t), \tag{65}$$

such that $\Delta\chi$ is small, of order V. In order that (59) be satisfied, we require

$$\mathrm{Tr}_R\, \Delta\chi(t) = 0, \tag{66}$$

so that

$$s(t) = \mathrm{Tr}_R\, \chi(t),$$

If we put (65) into the integrand of (64) and retain only terms of order V^2 on the right, we have

$$\frac{\partial s}{\partial t} = \frac{-1}{\hbar^2}\int_0^t \mathrm{Tr}_R\Big[V(t), [V(t'), s(t')f_0(R)]\Big]\,\mathrm{d}t'. \tag{67}$$

The reduced density operator $s(t)$ determines the statistical properties of the system. Since $s(t')$ occurs in the integrand, the behaviour of the system in the future depends on its past history from $t = 0$ to t. We next make the assumption that the system is Markoffian which means that its future behaviour is determined by the present and not the past. This assumption is valid, since damping destroys knowledge of the past. Mathematically, we therefore replace $s(t')$ by its present value $s(t)$ so that (67) reduces to

$$\frac{\partial s}{\partial t} = \frac{-1}{\hbar^2}\int_0^t \mathrm{Tr}_R\Big[V(t), [V(t'), s(t)f_0(R)]\Big]\,\mathrm{d}t' \tag{68}$$

which is called the master equation.

The Markoffian approximation has been done very crudely for the sake of brevity. Actually, one should take a coarse grained average on a time scale which is long compared with the reservoir correlation time but short compared with the system damping time. It should also be long enough to contain many cycles of the undamped system motion. We may then let the upper limit in (68) go to infinity.

Next we assume the interaction energy between system and reservoir can be written in the form

$$V = \sum_i F_i\, Q_i, \tag{69}$$

where the F_i are reservoir operators and the Q_i are system operators. In the interaction picture these may be written as

$$V(t) = \sum_i F_i(t) Q_i \exp(i\omega_i t) \tag{70}$$

When this is substituted into (68) and the double commutator is expanded, we find after minor algebraic manipulations that

$$\frac{\partial s}{\partial t} = -\sum_{i,j}\delta(\omega_i, -\omega_j)\,\{[Q_i\, Q_j\, s - Q_j\, s\, Q_i]w^+_{ij} - [Q_i\, s\, Q_j - s\, Q_j\, Q_i]w^-_{ji}\}, \tag{71}$$

where we have let

$$w_{ij}^{+} = \int_0^\infty \exp(-i\omega_j t)\,\mathrm{Tr}_R\,[f_0(R)\,F_i(t)\,F_j]\,\mathrm{d}t$$
$$w_{ji}^{-} = \int_0^\infty \exp(-i\omega_j t)\,\mathrm{Tr}_R[f_0(R)\,F_j\,F_i(t)]\,\mathrm{d}t. \tag{72}$$

This master equation describes the statistical behaviour of a system coupled to a reservoir under the Markoff approximation.

b. *Driven Damped Oscillator*

Let us apply the above analysis to obtain the master equation for the reduced density operator in the Schrödinger picture for a driven damped mode of the radiation field in a cavity. The unperturbed Hamiltonian is

$$H_0 = \hbar\omega_c\, a^\dagger a + \sum_j \hbar\omega_j\, b_j^\dagger\, b_j, \tag{73}$$

where ω_c is the cavity frequency.
We take the interaction energy to be

$$V = \hbar \sum_l (\kappa_l\, b_l\, a^\dagger + \kappa_l^*\, b_l^\dagger\, a) \tag{74}$$

where κ_l describes the scattering process.

In the notation of (70), we have

$$Q_1 = a^\dagger \qquad Q_2 = a = Q_1^\dagger$$
$$F_1 = \sum_l \kappa_l\, b_l \qquad F_2 = \sum_l \kappa_l^*\, b_l^\dagger = F_1^\dagger. \tag{75}$$

In the interaction picture we easily see that

$$Q_1^I = a^\dagger \exp[i\omega_c(t-t_0)] \qquad = Q_2^{\dagger I}$$
$$F_1^I = \sum_l \kappa_l\, b_l \exp[-i\omega_l(t-t_0)] \; = F_2^{\dagger I}. \tag{76}$$

Our first task is to compute the reservoir spectral densities w_{ij}^{+} and w_{ji}^{-}. We shall calculate one in detail and leave the others as an exercise. We have by (72) and (76),

$$w_{21}^{+} = \int_0^\infty \exp(i\omega_2\tau)\,\langle F_2(\tau)\,F_1\rangle_R\,\mathrm{d}\tau. \tag{77}$$

In the notation of (72), we see that

$$\omega_2 = -\omega_c. \tag{78}$$

Also by (75)

$$\langle F_2(\tau)\, F_1\rangle_R = \left\langle \sum_l \kappa_l^* \exp(i\omega_l \tau)\, b_l^\dagger \sum_m \kappa_m\, b_m \right\rangle_R$$
$$= \sum_{l,m} \kappa_l^*\, \kappa_m \exp(i\omega_l \tau)\, \langle b_l^\dagger b_m\rangle_R \tag{79}$$

But

$$\langle b_l^\dagger\, b_m\rangle_R = \frac{\mathrm{Tr}_R \exp(-\beta R)\, b_l^\dagger b_m}{\mathrm{Tr}_R \exp(-\beta R)}, \tag{80}$$

where $R = \sum_j \hbar\omega_j\, b_j\, b_j$. The reader may show easily that

$$\langle b_l^\dagger\, b_m\rangle_R = \delta_{lm}\, \bar{n}_l, \tag{81a}$$

where

$$\bar{n}_l = \frac{1}{\exp(\hbar\omega_l/kT)-1} \tag{81b}$$

Therefore, (77) becomes on using these results

$$w_{21}^+ = \sum_l |\kappa_l|^2\, \bar{n}_l \int_0^\infty \exp[i(\omega_l-\omega_c)\tau]\, \mathrm{d}\tau, \tag{82}$$

where we have interchanged the order of summation and integration. Let $g(\omega_l)\,\mathrm{d}\omega_l$ be the number of reservoir modes between ω_l and $\omega_l+\mathrm{d}\omega_l$, then since we assume that these modes are closely spaced, we may change the sum to an integral

$$\sum_l \{\} \to \int_0^\infty \mathrm{d}\omega_l\, g(\omega_l)\, \{\}, \tag{83}$$

so that (82) becomes

$$w_{21}^+ = \int_0^\infty \mathrm{d}\omega_l\, g(\omega_l)\, |\kappa(\omega_l)|^2\, \bar{n}(\omega_l)\, I, \tag{84a}$$

where we let

$$I = \int_0^\infty \exp[i(\omega_l-\omega_c)\tau]\, \mathrm{d}\tau. \tag{84b}$$

Now

$$\int_0^\infty \exp(\pm i\Omega\tau)\, \mathrm{d}\tau = \pi\delta(\Omega) \pm i\mathscr{P}\, \frac{1}{\Omega}, \tag{85}$$

where $\mathscr{P}$ is the Cauchy principle part defined by

$$\int_{-a}^{+b} \mathscr{P} \frac{f(\Omega)}{\Omega} d\Omega = \lim_{\varepsilon \to 0} \left\{ \int_{-a}^{-\varepsilon} \frac{f(\Omega)}{\Omega} d\Omega + \int_{\varepsilon}^{b} \frac{f(\Omega) d\Omega}{\Omega} \right\}. \tag{86}$$

Therefore, (84a) becomes

$$w_{21}^{+} = \int_0^\infty d\omega_l \, g(\omega_l) \, |\kappa(\omega_l)|^2 \, \bar{n}(\omega_l) \left\{ \pi \delta(\omega_l - \omega_c) + i \mathscr{P} \frac{1}{\omega_l - \omega_c} \right\}$$

$$\equiv \left(\frac{\gamma}{2} - i\Delta\omega \right) \bar{n}, \tag{87}$$

where we have let

$$\gamma = 2\pi \, g(\omega_c) \, |\kappa(\omega_c)|^2 \tag{88}$$

$$\Delta\omega = - \mathscr{P} \int_0^\infty \frac{d\omega_l \, g(\omega_l) \, |\kappa(\omega_l)|^2}{\omega_l - \omega_c}$$

and

$$\bar{n} = \frac{1}{\exp(\hbar\omega_c / kT) - 1}. \tag{89}$$

The imaginary part of w_{21}^{+} represents a small shift in the cavity frequency due to the coupling to the reservoir and in general may be neglected.

The remaining w's may be found by the same argument and are given by

$$w_{21}^{+} = \left(\frac{\gamma}{2} - i\Delta\omega \right) \bar{n} = w_{21}^{-*}$$

$$w_{12}^{+} = \left(\frac{\gamma}{2} + i\Delta\omega \right) (1 + \bar{n}) = w_{12}^{-*} \tag{90}$$

$$w_{11}^{\pm} = w_{22}^{\pm} = 0.$$

If we use these and (75) and (76) in (71), we find after minor algebra that

$$\frac{\partial S}{\partial t} = -i(\omega_c + \Delta\omega) [a^\dagger a, S] + \frac{\gamma}{2} [2aSa^\dagger - a^\dagger aS - Sa^\dagger a]$$

$$+ \gamma \bar{n} [a^\dagger Sa + aSa^\dagger - a^\dagger aS - Saa^\dagger]. \tag{91}$$

We see therefore, that the only effect of $\Delta\omega$ is to change slightly the cavity resonant frequency ω_c. $\Delta\omega$ is proportional to $|\kappa|^2$ by (88), which is the small coupling coefficient. We may re-define $\omega_c + \Delta\omega$ as ω_c' or neglect it. All operators in (91) are in the Schrödinger picture.

The first term in (91) represents the free motion of the cavity mode. The γ terms represent the loss of energy from the system to the reservoir, while the $\gamma\bar{n}$ terms represent the diffusion of fluctuations in the reservoir into the system mode. Note by (89) that as the reservoir approaches absolute zero, $\bar{n} \to 0$.

We may transform (91) to the interaction picture. If we let

$$H' = \hbar(\omega_c + \Delta\omega)\, a^\dagger a = \hbar\omega_c'\, a^\dagger a, \tag{92}$$

then (91) becomes in the interaction picture,

$$\frac{\partial s}{\partial t} = +\frac{\gamma}{2}[2asa^\dagger - a^\dagger as - sa^\dagger a] + \gamma\bar{n}[a^\dagger sa + asa^\dagger - a^\dagger as - saa^\dagger], \tag{93}$$

where all operators are in the interaction picture. We omitted the superscript I in the γ and $\gamma\bar{n}$ terms for simplicity since all these terms contain both an a and an $a^\dagger$ and we note that $a^I a^{\dagger I} = aa^\dagger$ and no confusion is likely to arise.

The master equation (93) may be solved by various techniques.

3. Quantum-Classical Correspondence[2]

The essential mathematical difference between quantum and classical mechanics is the algebra obeyed. In quantum mechanics the variables do not necessarily commute and the order in which they are written down is important. By introducing the concept of ordered operators, we may set up a correspondence between the quantum system and a "classical" equivalent system. This allows us to separate the stochastic problem from the quantum problem.

3.1. Ordered Operators—Distribution Function

Let us consider a complete set of non-commuting operators $a_1, a_2, \ldots, a_f$ in the Schrödinger picture which obey some set of commutation or anticommutation relations. The anticommutator of two operators is defined by $[a_i, a_j]_+ \equiv a_i a_j + a_j a_i$. Suppose we have some function Q of these operators which may be expanded in a power series. We may use the commutation (or anticommutation) relations to re-order the terms in the function into some pre-determined or chosen order. Let this chosen order be $a_1, a_2, a_3, \ldots, a_f$. Therefore, Q in chosen order becomes

$$Q = Q^c(a_1 \ldots, a_f) = \sum_{r_1} \cdots \sum_{r_f} Q^c_{r_1, \ldots, r_f}\, a_1^{r_1} a_2^{r_2} \cdots a_f^{r_f}. \tag{94}$$

We put a superscript c to indicate that we have put the function in chosen order which of course is equal to the function in the original order. Once we

have put Q in chosen order, we may define an associated c-number function by means of

$$\bar{Q}^c(\alpha_1, \alpha_2, \ldots, \alpha_f) = \sum_{r_1} \cdots \sum_{r_f} Q^c_{r_1, r_2, \ldots, r_f} \alpha_1^{r_1} \alpha_2^{r_2} \cdots \alpha_f^{r_f}, \tag{95}$$

where we replace the operator a_i by the c-number α_i which is real or complex depending on whether a_i is Hermitian or not. We put a bar to remind us we are now dealing with a quasi-classical or c-number function.

We next define a linear chosen ordering operator $\mathscr{C}$ by means of

$$Q^c(a_1, a_2, \ldots, a_f) = \mathscr{C}\{\bar{Q}^c(\alpha_1, \ldots \alpha_f)\}, \tag{96}$$

where $\mathscr{C}$ tells us to replace each α_i by the corresponding operator and write all terms in chosen order. We may also define the inverse operator $\mathscr{C}^{-1}$.

We may give a formal representation of the $\mathscr{C}$ operator by means of

$$\mathscr{C}\{\bar{Q}^c(\alpha_1, \ldots, \alpha_f)\} = \int \cdots \int \bar{Q}^c(\alpha_1, \ldots, \alpha_f) \prod_{i=1}^{f} \delta(\alpha_i - a_i)\, \mathrm{d}\alpha_i, \tag{97}$$

where the δ-functions are operators *in the chosen order*. In the integration we formally replace each α_i by the corresponding a_i and put all terms in the chosen order. But since this is exactly what $\mathscr{C}$ tells us to do, the two expressions in (97) are formally equivalent.

If the operator a_i is Hermitian and the α_i real, we may represent the δ-function by means of

$$\delta(\alpha - a) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \exp\left[-i\xi(\alpha - a)\right] \mathrm{d}\xi, \tag{98}$$

while if it is non-Hermitian, the operator and its adjoint will both be present and we then let

$$\delta(\alpha - a)\, \delta(\alpha^* - a^\dagger) = \frac{1}{\pi^2} \int \int \exp\left[-i\xi(\alpha - a)\right] \exp\left[-i\xi^*(\alpha^* - a^\dagger)\right] \mathrm{d}^2\xi, \tag{99}$$

if the chosen order is a, $a^\dagger$. Here we integrate over the entire complex ξ-plane and $\mathrm{d}^2\xi = \mathrm{d}(\mathrm{Re}\,\xi)\, \mathrm{d}(\mathrm{Im}\,\xi)$. In this formal presentation we shall use (98) for simplicity.

If we use (98), (97) may be rewritten as

$$Q^c(a_1, \ldots, a_f) = \left(\frac{1}{2\pi}\right)^f \int \cdots \int \mathrm{d}\alpha_1 \ldots \mathrm{d}\alpha_f\, \bar{Q}^c(\alpha_1, \ldots, \alpha_f)$$
$$\times \int \cdots \int \mathrm{d}\xi_1 \ldots \mathrm{d}\xi_f \exp\left[-i\xi_1(\alpha_1 - a_1)\right] \ldots \exp\left[-i\xi_f(\alpha_f - a_f)\right]. \tag{100}$$

If we interchange the order of integration of the ξ's and α's, we may write this as

$$Q^c(a_1, ..., a_f) = \int ... \int d\xi_1 ... d\xi_f \exp(i\xi_1 a_1) ... \exp(i\xi_f a_f) F(\xi_1, ..., \xi_f), \tag{101a}$$

where

$$F(\xi_1, ..., \xi_f) = \left(\frac{1}{2\pi}\right)^f \int ... \int d\alpha_1 ... d\alpha_f \exp[-i(\xi_1\alpha_1 + ... + \xi_f\alpha_f) \times \bar{Q}^c(\alpha_1, ..., \alpha_f). \tag{101b}$$

Therefore, $(F\xi_1, ..., \xi_f)$ is just the Fourier transform of the associated c-number function $\bar{Q}^c(\alpha_1, ..., \alpha_f)$. If we invert (101b) by taking the inverse Fourier transform, we obtain

$$\bar{Q}^c(\alpha_1, ..., \alpha_f) = \int ... \int \exp[i(\xi_1\alpha_1 + ... + \xi_f\alpha_f)] F(\xi_1, ..., \xi_f) d\xi_1 ... d\xi_i, \tag{101c}$$

which gives us the associated function in terms of F. If we apply the $\mathscr{C}$ operator to both sides of (101c), we obtain (101a) exactly.

Consider next the expectation value of Q

$$\langle Q^c(a_1, ..., a_f, t)\rangle = \mathrm{Tr}\, \rho(t)\, Q^c(a_1, ..., a_f). \tag{102}$$

If we use (101a), we have

$$\langle Q^c(a_1, ..., a_f, t)\rangle = \int ... \int d\xi_1 ... d\xi_f\, C(\xi_1, ..., \xi_f, t)\, F(\xi_1, ..., \xi_f), \tag{103}$$

where

$$C(\xi_1, ..., \xi_f, t) = \langle \exp(i\xi_1 a_1) ... \exp(i\xi_f a_f)\rangle = \mathrm{Tr}\,[\rho(t) \exp(i\xi_1 a_1) ... \exp(i\xi_f a_f)] \tag{104}$$

is a characteristic function and $F(\xi_1, ...\xi_f)$ is given by (101b) and is the Fourier transform of the associated function $\bar{Q}^c(\alpha_1, ..., \alpha_f)$. In (103) we have converted the quantum expectation value to an integral over c-number functions. We may express this in another way as follows. By (97) we have an alternate expression equivalent to (103)

$$\langle Q^c(a_1, ..., a_f, t)\rangle = \int ... \int d\alpha_1 ... d\alpha_f\, \bar{Q}^c(\alpha_1, ..., \alpha_f) P_c(\alpha_1, ..., \alpha_f, t), \tag{105}$$

where we have let

$$P_c(\alpha_1, ..., \alpha_f, t) = \langle \delta(\alpha_1 - a_1) ... \delta(\alpha_f - a_f)\rangle = \mathrm{Tr}\, \rho(t)\, \delta(\alpha_1 - a_1) ... \delta(\alpha_f - a_f)$$

$$= \left(\frac{1}{2\pi}\right)^f \int ... \int d\xi_1 ... d\xi_f \exp[-i(\xi_1\alpha_1 + ... + \xi_f\alpha_f)\, C(\xi_1, ..., \xi_f, t), \tag{106}$$

and where we have used (98) and (104). We call P a distribution function which is the Fourier transform of the characteristic function. If we apply the inverse Fourier transform to (106), we have

$$C(\xi_1, \ldots, \xi_f, t) = \int \ldots \int d\alpha_1 \ldots d\alpha_f \exp\left(i[\xi_1\alpha_1 + \ldots + \xi_f \alpha_f)\right] P_c(\alpha_1, \ldots, \alpha_f, t)$$
$$= \mathrm{Tr} \exp(i\xi_1 a_1) \ldots \exp(i\xi_f a_f)\, \rho(a_1, \ldots, a_f, t). \tag{107}$$

The advantage of using the distribution function stems from the fact that expectation values of operators in chosen order are evaluated just like classical averages if P_c is considered as a classical probability. We may see this as follows. We have by (107)

$$\left.\frac{\partial^{r_1+r_2+\ldots r_f} C}{\partial(i\xi_1)^{r_1}\, \partial(i\xi_2)^{r_2} \ldots \partial(i\xi_f)^{r_f}}\right|_{\xi_i = 0} = \langle a_1^{r_1} a_2^{r_2} \ldots a_f^{r_f} \rangle$$
$$= \int \ldots \int \alpha_1^{r_1}\, \alpha_2^{r_2} \ldots \alpha_f^{r_f} P_c(\alpha_1, \ldots, \alpha_f, t)\, \mathrm{d}\alpha_1 \ldots \mathrm{d}\alpha_f. \tag{108}$$

There will be corrections to this quantum-classical correspondence for mean values of operators which are not in chosen order. For example.

$$\langle a_2\, a_1 \rangle \neq \int\int \alpha_1\, \alpha_2\, P_c(\alpha_1, \alpha_2, t)\, \mathrm{d}\alpha_1\, \mathrm{d}\alpha_2 = \langle a_1\, a_2 \rangle, \tag{109}$$

if a_1, a_2 is the chosen order.

3.2. EQUATION OF MOTION FOR DISTRIBUTION STUDY[3]

We may next use the above association scheme in order to transform the master operator equation (71) into an equation of motion for the c-number distribution function. From the definition (106), we have

$$\frac{\partial P_c}{\partial t}(\boldsymbol{\alpha}, t) = \mathrm{Tr} \frac{\partial S}{\partial t} \delta^c(\boldsymbol{\alpha} - \mathbf{a}), \tag{110}$$

where we let

$$\delta^c(\boldsymbol{\alpha} - \mathbf{a}) = \prod_{i=1}^{f} \delta(\alpha_i - a_i). \tag{111}$$

If we use (71) in the Schrödinger picture and the cyclic property of traces, (110) reduces to

$$\frac{\partial P_c}{\partial t} = \mathrm{Tr}\, S \left\{ \frac{1}{i\hbar} [\delta^c(\boldsymbol{\alpha} - \mathbf{a}), H] \right.$$
$$\left. - \sum_{i,j} \delta(\omega_i, -\omega_j) \left\{ [\delta^c(\boldsymbol{\alpha} - \mathbf{a}), Q_i]\, Q_j\, w_{ij}^{+} - Q_j [\delta^c(\boldsymbol{\alpha} - \mathbf{a}), Q_i] w_{ji}^{-} \right\} \right\}. \tag{112}$$

We may represent the δ-function as

$$\delta^c(\alpha-\mathbf{a}) = \prod_i \exp\left(-a_i \frac{\partial}{\partial \alpha_i}\right) \delta(\alpha_i) = \exp\left[-\mathbf{a}\frac{\partial}{\partial \alpha}\right] \delta(\alpha) \tag{113}$$

and (112) becomes

$$\frac{\partial P_c}{\partial t} = \mathrm{Tr}\, S \left\{ \frac{1}{i\hbar}\left[\exp\left(-\mathbf{a}\frac{\partial}{\partial \alpha}\right), H\right] - \sum_{i,j} \delta(\omega_i, -\omega_j) \right.$$

$$\left. \times \left\{ \left[\exp\left(-\mathbf{a}\frac{\partial}{\partial \alpha}\right), Q_i\right] Q_j w_{ij}^{+} - Q_j \left[\exp\left(-\mathbf{a}\frac{\partial}{\partial \alpha}\right), Q_i\right] w_{ji}^{-} \right\} \right\} \delta(\alpha)$$

$$= \mathrm{Tr}\, S L' \left(\frac{\partial}{\partial \alpha}, \mathbf{a}\right) \delta(\alpha), \tag{114}$$

where L' stands for the operator argument of the trace which is a function of the system operators $a_1, \ldots, a_f$. The Q_i and Q_j are usually one of these, or functions of them. Since L' is a function of the a's, we may in principle use the commutation relations to put it into the chosen c-order and

$$L'\left(\frac{\partial}{\partial \alpha}, \mathbf{a}\right) = L'^c\left(\frac{\partial}{\partial \alpha}, \mathbf{a}\right) = \int \bar{L}'^c\left(\frac{\partial}{\partial \alpha}, \boldsymbol{\beta}\right) \delta^c(\boldsymbol{\beta}-\mathbf{a})\, \mathrm{d}\boldsymbol{\beta}, \tag{115}$$

where we have used (97) to express L'. Now by inspection of L' in (114), we see that after putting it in chosen order, the exponentials in $\bar{L}'^c\{\partial/\partial\alpha, \boldsymbol{\beta}\}$ can be factorized out of all terms, so we may write it as

$$\bar{L}'^c\left(\frac{\partial}{\partial \alpha}, \boldsymbol{\beta}\right) = \bar{L}^c\left(\frac{\partial}{\partial \alpha}, \boldsymbol{\beta}\right) \exp\left(-\boldsymbol{\beta}\frac{\partial}{\partial \alpha}\right). \tag{116}$$

When we use this and (115), (114) becomes

$$\frac{\partial P_c}{\partial t}(\alpha, t) = \mathrm{Tr}\, S \int \bar{L}^c\left(\frac{\partial}{\partial \alpha}, \boldsymbol{\beta}\right) \exp\left(-\boldsymbol{\beta}\frac{\partial}{\partial \alpha}\right) \delta(\alpha)\, \delta^c(\boldsymbol{\beta}-\mathbf{a})\, \mathrm{d}\boldsymbol{\beta}. \tag{117}$$

However,

$$\exp\left(-\boldsymbol{\beta}\frac{\partial}{\partial \alpha}\right) \delta(\alpha) = \delta(\boldsymbol{\beta}-\alpha), \tag{118}$$

so that (117) becomes

$$\frac{\partial P_c}{\partial t}(\alpha, t) = \mathrm{Tr}\, S \int \bar{L}^c\left(\frac{\partial}{\partial \alpha}, \boldsymbol{\beta}\right) \delta(\boldsymbol{\beta}-\alpha)\, \delta^c(\boldsymbol{\beta}-\mathbf{a})\, \mathrm{d}\boldsymbol{\beta}$$

$$= \int \bar{L}^c\left(\frac{\partial}{\partial \alpha}, \boldsymbol{\beta}\right) \delta(\boldsymbol{\beta}-\mathbf{a})\, P_c(\boldsymbol{\beta}, t)\, \mathrm{d}\boldsymbol{\beta}, \tag{119}$$

where we used the definition (106) for P_c. If we carry out the β integration, we obtain

$$\frac{\partial P_c}{\partial t}(\alpha, t) = \bar{L}^c\left(\frac{\partial}{\partial \alpha}, \alpha\right) P_c(\alpha, t). \tag{120}$$

This is the formal equation obeyed by the distribution function which we wished to obtain. In many cases of interest, in particular for a homogeneously broadened single mode laser, we may neglect derivatives higher than second order in (120) and write it in the form

$$\frac{\partial P_c}{\partial t}(\alpha, t) = -\frac{\partial}{\partial \alpha_i}[\mathscr{A}_i(\alpha) P_c(\alpha, t)] + \frac{\partial}{\partial \alpha_i \partial \alpha_j}[\mathscr{D}_{ij}(\alpha) P_c(\alpha, t)], \tag{121}$$

where we sum i and j from 1 to f. This is called a Fokker–Planck equation and describes the statistical behaviour of an associated classical stochastic process. The $\mathscr{A}_i$'s form the components of a drift vector which give the mean motion of a_i as we shall show next. We have by (105)

$$\frac{\mathrm{d}\langle a_i\rangle}{\mathrm{d}t} = \int \frac{\partial P_c}{\partial t}(\alpha, t)\, \alpha_i \, \mathrm{d}\alpha. \tag{122}$$

If we put (121) into this and integrate by parts once on the first term and by parts twice on the second, we obtain

$$\frac{\mathrm{d}\langle a_i\rangle}{\mathrm{d}t} = \int \mathscr{A}_i(\alpha, t) P_c(\alpha, t)\, \mathrm{d}\alpha, \tag{123}$$

since the integrated parts vanish.

The terms containing $\mathscr{D}_{ij}$ represent diffusion. The $\mathscr{D}_{ij}$'s are called diffusion coefficients and form the elements of a diffusion matrix. Consider two operators $a_i a_j$ and assume ij is in chosen order.

We then have

$$\frac{\mathrm{d}}{\mathrm{d}t} a_i a_j = \int \frac{\partial P_c}{\partial t} \alpha_i \alpha_j \, \mathrm{d}\alpha. \tag{124}$$

If we again use (121) and integrate by parts, we obtain

$$\frac{\mathrm{d}}{\mathrm{d}t} a_i a_j = \int P_c(\alpha, t)\, \mathrm{d}\alpha \{\alpha_i \mathscr{A}_j + \alpha_j \mathscr{A}_i + 2\mathscr{D}_{ij}\}, \tag{125}$$

which shows how the diffusion affects the motion.

3.3. TWO-TIME AVERAGES. SPECTRA[3]

A complete statistical description requires mean values of operators at different times. For example, in the study of the statistical properties of a mode of a radiation field, the fluctuation spectrum is defined by

$$\int_{-\infty}^{\infty} \exp(-i\omega t)\langle a^\dagger(t)\, a(0)\rangle \, \mathrm{d}t. \tag{126}$$

The intensity spectrum is given by

$$\int_{-\infty}^{\infty} \exp(-i\omega t)\langle a^\dagger(0)\, a^\dagger(t)\, a(t)\, a(0)\rangle \, \mathrm{d}t, \tag{127}$$

while the photon number spectrum is

$$\int_{-\infty}^{\infty} \exp(-i\omega t)\langle a^\dagger(t)\, a(t)\, a^\dagger(0)\, a(0)\rangle \, \mathrm{d}t. \tag{128}$$

These are directly related to experimentally observable quantities which the theory must be capable of predicting. In the interests of brevity we shall merely give the results of such averages.

Let $P_c(\boldsymbol{\alpha} t \mid \boldsymbol{\beta}, 0)$ be the solution of the equation of motion for the distribution function subject to the initial condition that $\alpha_i = \beta_i$. Then it can be shown that

$$\langle M(t) N(0)\rangle = \int \mathrm{d}\boldsymbol{\alpha} \int \mathrm{d}\boldsymbol{\beta}\, \overline{M}^c(\boldsymbol{\alpha})\, P_c(\boldsymbol{\alpha}, t \mid \boldsymbol{\beta}, 0)\, \mathrm{Tr}[S(0)\, \delta^c(\boldsymbol{\beta} - \mathbf{a})\, N(0)], \tag{129}$$

where M and N are two-system operators and $S(0)$ is the initial reduced system density operator.

4. ROTATING WAVE VAN DER POL OSCILLATOR

It has been shown[4] that a single mode homogeneously broadened laser satisfies the Fokker–Planck equation for a rotating wave van der Pol oscillator given by

$$\frac{\partial P_n}{\partial t}(\alpha, \alpha^*, t) = -\frac{\partial}{\partial \alpha}(g - i\omega_c - |\alpha|^2)\alpha P_n - \frac{\partial}{\partial \alpha^*}(g + i\omega_c - |\alpha|^2)\alpha^* P_n + 4\frac{\partial^2 P_n}{\partial \alpha \partial \alpha^*}, \tag{130}$$

where g is a numerical pumping parameter and the equation has been scaled in the time and α variables. The chosen order is normal order. It describes a

radiation field mode which is damped for $g<0$ and starts to grow exponentially when $g>0$. $g=0$ is called the oscillation threshold. For $g>0$ the oscillation builds up but because of the nonlinear terms in $|\alpha|^2\alpha$ and $|\alpha|^2\alpha^*$, the oscillations will stabilize at a fixed amplitude, since $g-|\alpha|^2$ eventually goes to zero as $|\alpha|^2$ increases.

Because of the $|\alpha|^2$ terms, we cannot solve (130) analytically. However, there are two interesting regimes where we may obtain analytical solutions, viz., far below threshold ($|\alpha|^2 \ll |g|$) and far above threshold ($|\alpha|^2 \gg |g|$).

(a) *Far Below Threshold* ($|\alpha|^2 \ll |g|$)

In this region (130) reduces to

$$\frac{\partial P}{\partial t} = -\frac{\partial}{\partial \alpha}(g\alpha P) - \frac{\partial}{\partial \alpha^*}(g\alpha^* P) + 4\frac{\partial^2 P}{\partial \alpha \partial \alpha^*}. \tag{131}$$

The reader may easily verify that the Green's function solution is

$$P(\alpha, t\,|\,\beta, 0) = \frac{1}{\pi\xi(t)} \exp\left\{-\frac{|\alpha - \beta \exp[-(i\omega_c - g)t]|^2}{\xi(t)}\right\}, \tag{132}$$

where

$$\xi(t) = \frac{2}{g}[\exp(2gt) - 1] \tag{133}$$

and $g<0$ (below threshold). We assume at $t=0$ the system is in a steady state given by the Gaussian

$$P(\beta, 0) = \frac{|g|}{2\pi} \exp\left(-\frac{|g|}{2}|\beta|^2\right). \tag{134}$$

The fluctuation spectrum below threshold is seen to be Lorentzian with half width $|g|$ and is given by

$$\int_{-\infty}^{\infty} \langle a^\dagger(t)\, a(0)\rangle \exp(i\omega t)\, \mathrm{d}t = \frac{4}{(\omega_c - \omega)^2 + g^2}. \tag{135}$$

(b) *Far Above Threshold* $|\alpha|^2 \gg |g|$

To study the Fokker–Planck equation above threshold, it is best to change to polar coordinates and let

$$\alpha = r \exp[i(\phi - \omega_c t)]\, \mathrm{d}^2\alpha = r\, \mathrm{d}r\, \mathrm{d}\phi. \tag{136}$$

Then (130) becomes

$$\frac{\partial P}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial P}{\partial r}\right) + \frac{1}{r^2}\frac{\partial^2 P}{\partial \phi^2} - \frac{1}{r}\frac{\partial}{\partial r}[(g-r^2)r^2 P] \tag{137}$$

and P is normalized so that

$$\int_0^{2\pi} \mathrm{d}\phi \int_0^{\infty} P(r,\phi,t)\, r\, \mathrm{d}r = 1. \tag{138}$$

In the steady state, P is independent of t and the ϕ-independent solution is

$$P_s(r) = \frac{\exp -\frac{1}{4}(r^2-g)^2}{\int_{-g/2}^{\infty} \exp(-x^2)\, \mathrm{d}x}, \tag{139}$$

The mean number of photons in the steady state is given by

$$\langle a^\dagger a\rangle_s = \int_0^{\infty} r^2 P_s(r)\, r\, \mathrm{d}r. \tag{140}$$

Far above threshold we may let the lower limit in (139) go to $-\infty$ so that

$$\langle a^\dagger a\rangle_s \approx g. \tag{141}$$

To study the fluctuations about this steady state we let

$$r = \sqrt{g} + r_1(t), \tag{142}$$

where r_1 is small. If we use this and let

$$P(r,\phi,t) = \frac{R(r_1,t)\,\Phi(\phi,t)}{\sqrt{g}}, \tag{143}$$

(137) separates into

$$\begin{aligned} \frac{\partial R}{\partial t} &= \frac{\partial^2 R}{\partial {r_1}^2} + \frac{\partial}{\partial r_1}(2g r_1 R) \\ \frac{\partial \Phi}{\partial t} &= \frac{1}{g}\frac{\partial^2 \Phi}{\partial \phi^2}, \end{aligned} \tag{144}$$

The steady state solutions are

$$\begin{aligned} R_s &= \sqrt{\frac{g}{\pi}} \exp(-g{r_1}^2) \\ \Phi_s &= \frac{1}{2\pi}, \end{aligned} \tag{145}$$

so the amplitude has a Gaussian distribution and the phase is completely random. The non-steady state Green's function solution of (144) is

$$P(\alpha, t \mid \beta, 0) = \frac{1}{\sqrt{[\pi g \xi(t)]}} \exp\left\{\frac{-[r_1 - \eta(t)]^2}{\xi(t)}\right\} \times \frac{1}{2\pi} \sum_{m=-\infty}^{\infty} \exp\left\{in(\phi - \phi_0 + \omega_c t) - \frac{n^2}{g} t\right\}, \quad (146)$$

where

$$\xi(t) = \frac{1}{g}[1 - \exp(-4gt)] \quad (147)$$

$$\eta(t) = r_{10} \exp(-2gt).$$

The fluctuation spectrum is given by

$$\int_{-\infty}^{\infty} \exp(-i\omega t)\langle a^\dagger(t)\, a(0)\rangle \, dt = \frac{2}{(\omega_c - \omega)^2 + (1/g)^2} + \frac{2 + (1/g^2)}{(\omega_c - \omega)^2 + [(1/g) + 2g]^2}, \quad (148)$$

which is the sum of two Lorentzians. The first has half width g^{-1} and is very narrow, and the second has half width $2g$ and is very broad.

References

1. Louisell, W. H., "Radiation and Noise in Quantum Electronics", McGraw–Hill Book Co., New York, (1964).
2. Lax, M., *Phys. Rev.* **172,** 350 (1968).
3. Louisell, W. H. and Marburger, J. H., *Phys. Rev.* **186,** 174 (1969).
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The Semiclassical and Quantum Theory of the Laser

H. HAKEN

For the purpose of this Summer School we have put the physics rather than the mathematics into the foreground. A physicist, who wants to learn about laser theory, usually first wishes to know the gross-features, and only afterwards does he want to learn about the more refined aspects like fluctuations With this in mind, we shall discuss the following:

1. *The structure of laser theory*
2. *Rate equations*
 Heuristic derivation
 Treatment of typical cases
3. *Semiclassical equations*
 Heuristic derivation
 How to come back to rate equations
 Treatment of typical cases
4. *Fully quantum mechanical equations*
 Derivation from first principles
 Some different approaches
 Treatment of typical cases
5. *General quantum theory of damping*

Readers who wish to make a quick survey of laser theory may read the following sections;

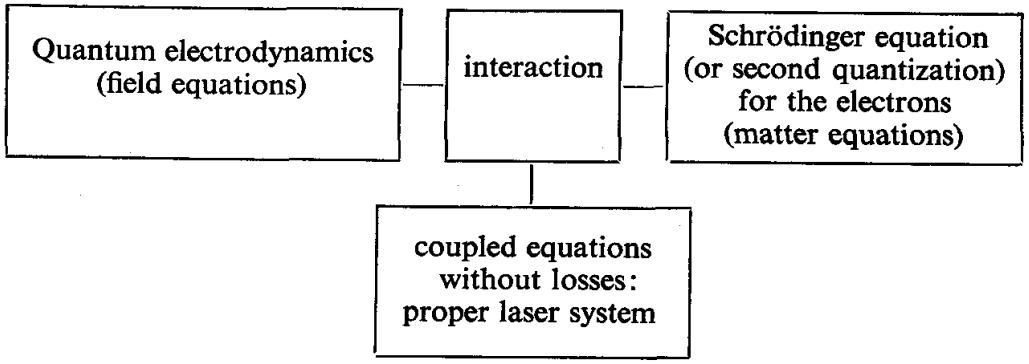
2.1–2.3; 3.1–3.7; (to equation 140); 4.1–4.3/4.5; 4.12.c; 4.12.d; 4.13.

These sections are written such that they can be read without knowledge of the omitted ones.

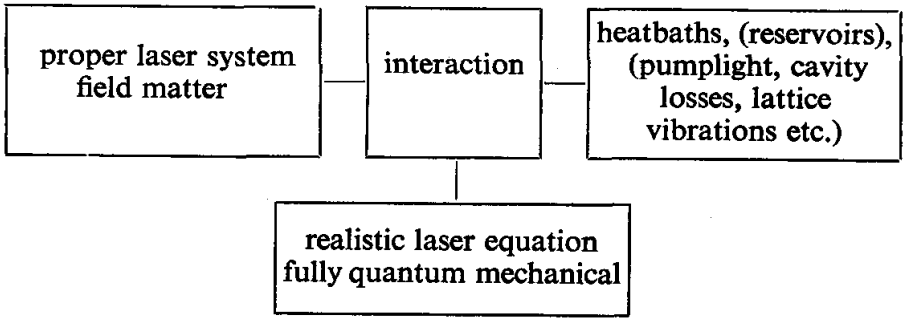
1. THE STRUCTURE OF LASER THEORY

Laser light is generated by stimulated emission (as it is expressed by the word laser). Both this process and spontaneous emission of light (in conventional light sources) are typical quantum mechanical effects. A proper laser theory must therefore be a quantum theory. Because light is an electro-magnetic field, the fundamental equations must be taken from quantum-electrodynamics. The emission processes are caused by the electrons of atoms (or solid

bodies), thus we have to consider the interaction between the quantized electromagnetic field (in short: field) and a system of electrons, which must be treated quantum mechanically, of course, in order for the theory to be consistent.

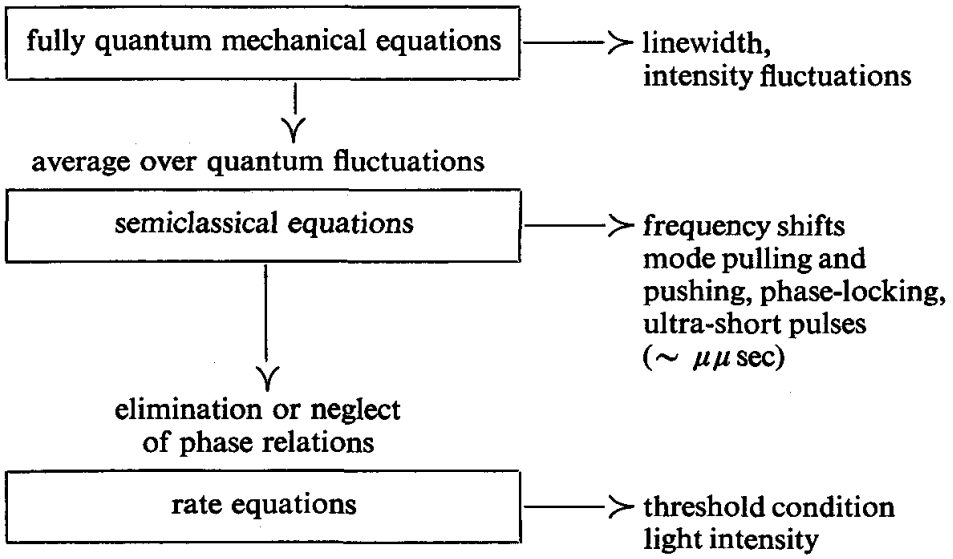


The field equations coupled with the matter equations give the basic set for the treatment of all kinds of processes in which light and matter (as far as the atomic shell is concerned) are involved. These equations are, however, insufficient for a laser theory, because they neglect the effect of the surroundings —the field can partly escape through the mirrors or may be absorbed by impurities etc.; the electrons interact with lattice vibrations, the pump-light, etc. All these effects stem from external reservoirs (or heatbaths), and are to a great extent of a statistical nature. Because laser action can be only maintained if the energy losses are balanced by the energy pumped into the system, it is quite clear, that any useful laser theory must incorporate these statistical effects.



A possible way of arranging the present lectures would be to derive the realistic laser equations from the very beginning and then to discuss their solutions.[1] We have, however, chosen a different way because the realistic and complete laser equations are, in general, rather complicated. On the other hand a number of gross-features of laser light can be understood by much simpler equations. This may be justified by asking the question as to what we expect from a laser theory. Firstly, we want to know the conditions under which laser action occurs, and the intensity of the laser light produced. The next question perhaps would concern the actual frequency of the laser light.

Finally we observe that the frequency is not completely sharp but has a finite width due to quantum fluctuations. A closer inspection of the complete laser equations reveals that their full content of information is needed only for the last question. To treat the second question it suffices to use equations which are averages of the fully quantum mechanical equations. These equations are called the semiclassical equations. Finally, the first question concerns only intensities, or, photon numbers, so that in general it is sufficient to discard phase relations and to use the so called rate equations for intensities. Thus we have the following scheme.



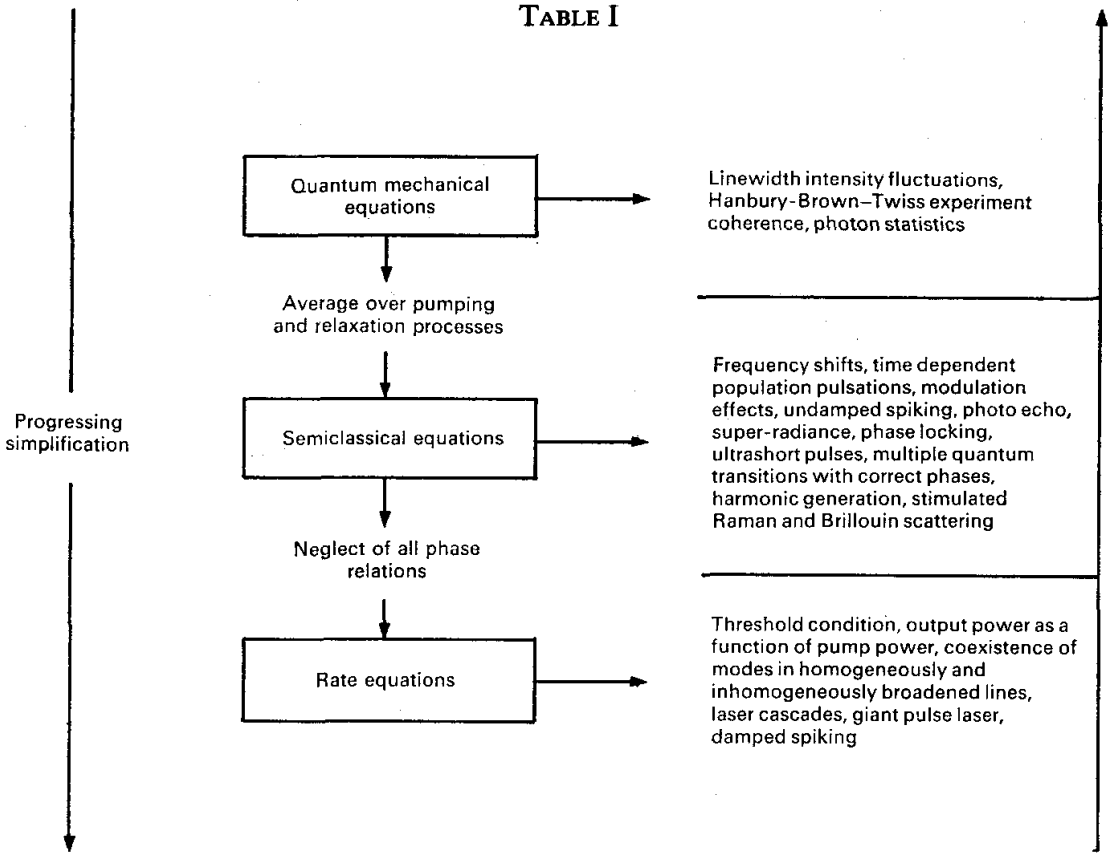
For a more complete survey consult Table 1

2. Rate Equations[2]

2.1. Equation for the Photon Number: Laser Condition

We all know from the usual quantum mechanics of particles that there exists a dualism between particle aspects (momentum, position) and wave aspects (frequency, wavelength). Similarly, such a dualism exists between photons and waves in quantum electrodynamics. The use of rate equations rests on the possibility of describing (at least under certain conditions discussed below) the light field in the photon picture. Following Einstein, we can define generation and annihilation rates of photons and thus establish rate equations. Let us consider a single mode with a photon number n, which is treated as a continuous variable. Its rate of change $\mathrm{d}n/\mathrm{d}t$, is caused by a number of different effects.

TABLE I



Emission and absorption of photons

Each excited atom *spontaneously* emits photons into the mode under consideration at a rate W. For simplicity we assume that this rate does neither depend on the position nor frequency of the individual atoms. This assumption is correct if the mode has infinite wavelength or is represented by a travelling wave (W : space-independent) and if the atomic line is homogeneously broadened (W : no individual frequency occurs).

If $N_{2,\mu}$ is the occupation number of the atom μ in its excited state, the total rate of spontaneously emitted photons is

$$\sum_{\mu=1}^{N} W N_{2,\mu}, \tag{1}$$

where the sum runs over all N atoms. Phase relations do not appear. The photon generation rate by *stimulated emission* is obtained from (1) by multiplying it with the photon number n:

$$\sum_{\mu=1}^{N} W N_{2,\mu} n. \tag{2}$$

On the other hand, the rate of *stimulated absorption is* proportional to the same rate W, to n, and to the number $\sum_{\mu=1}^{N} N_{1,\mu}$ of atoms in the lower state (1) and is given by

$$-\sum_{\mu=1}^{N} W N_{1,\mu}\, n. \tag{3}$$

The net gain of the stimulated process is thus

$$\sum_{\mu=1}^{N} W(N_2 - N_1)_\mu\, n. \tag{4}$$

Losses. The photons may escape from the cavity through the mirrors, by scattering processes, etc. Their decay rate is proportional to the number of photons present and is given by

$$-2\kappa n, \tag{5}$$

where κ is the cavity halfwidth. Putting the net gain (4) and the losses (5) together we find the photon rate equation:

$$\mathrm{d}n/\mathrm{d}t = -2\kappa n + \sum_{\mu=1}^{N} W(N_2 - N_1)_\mu\, n + \sum_{\mu=1}^{N} W N_{2,\mu}. \tag{6}$$

It always appears strange, that (6) contains no phase relations at all. We shall discuss this problem below. Because W does not depend on μ, we may introduce total occupation numbers

$$N_{j,\mathrm{tot}} = \sum_{\mu=1}^{N} N_{j,\mu}\,; \quad j = 1, 2, \tag{7}$$

and the total inversion ("population difference")

$$D = N_{2,\mathrm{tot}} - N_{1,\mathrm{tot}} = \sum_{\mu=1}^{N} (N_2 - N_1)_\mu. \tag{8}$$

With (7) and (8), the eqn (6) takes the form

$$\mathrm{d}n/\mathrm{d}t = -2\kappa n + D\, W n + N_{2,\mathrm{tot}} W. \tag{9}$$

It is customary to neglect the rate of spontaneously emitted photons because it is much smaller than the stimulated emission rate. Actually, there is a deeper reason for neglecting $W N_{2,\mathrm{tot}}$ compared to DWn and this will be treated in Section 4.

Equation (9) allows us to derive a laser condition. Laser action takes place if the generation rate is bigger than the loss rate, i.e. if

$$W \,.\, D > 2\kappa,$$

or

$$W(N_2 - N_1)_{\text{tot}} > 2\kappa. \tag{10}$$

Thus for laser action to take place, the inversion $D = (N_2 - N_1)_{\text{tot}}$ must be made sufficiently high. The photon life-time, t_1, in the cavity is related to 2κ by

$$2\kappa = 1/t_1. \tag{11}$$

We now derive an expression for W. Let the radiative lifetime of an atom be τ, so that it emits $1/\tau$ photons per second spontaneously. We are, however, interested only in one special "mode". We have therefore still to divide by p, the number of modes within the spontaneous linewidth $\Delta\nu$ of the atom. The number of photons spontaneously emitted per atom per second into a given mode is thus

$$W = 1/\tau p \tag{12}$$

For a Lorentzian line, p is given by† $8\pi^2 \nu^2 V \Delta\nu / c^3$ so that one obtains the laser condition[3]

$$\frac{(N_2 - N_1)_{\text{tot}}\, c^3}{8\tau\, \pi^2\, \nu^2 V \Delta\nu} > 2\kappa, \tag{13}$$

where V is the volume of the cavity and c is the velocity of light. In order to satisfy (13) the following conditions must be fulfilled.

(a) *The lifetime must be large enough.* This requires a detailed study of the losses of decaying modes in the Fabry–Perot resonators as well as in other resonator types. Also other loss mechanisms as well should be considered, e.g. impurity scattering, or coupling between modes.

(b) *The atomic linewidth* $\Delta\nu$ *must be small.* This is mainly a question of selecting materials but nevertheless requires a theoretical investigation of broadening mechanisms. Note that $\Delta\nu$ can be much bigger than $(1/\tau)$, because $\Delta\nu$ includes all kinds of broadening, e.g. lattice-vibrations, etc.

(c) *The inversion* $(N_2 - N_1)$ *must be high enough.* This requires a detailed knowledge of excitation mechanisms, e.g. optical pumping, etc.

† Here ν is the frequency defined as usual. Note, however, that in the remainder of the chapters, ν denotes the *circular* atomic frequency.

2.2. THE MATTER EQUATIONS

Because of spontaneous and stimulated emission, the *atomic occupation numbers* change. To find an equilibrium the atoms must be continuously pumped into their upper states. We first treat *two-level* atoms, and describe these processes by the following two equations for the single atom μ

$$dN_{2,\mu}/dt = -N_{2,\mu} w_{21} + N_{1,\mu} w_{12} - (N_2 - N_1)_\mu W n, \tag{14}$$

$$dN_{1,\mu}/dt = N_{2,\mu} w_{21} - w_{12} N_{1,\mu} + (N_2 - N_1)_\mu W n, \tag{15}$$

where the w's are transition rates.

The external pump is contained in w_{12}, and w_{21} contains both spontaneous emission into all modes and radiationless transitions. In practice the pumping is achieved via a third level which decays very rapidly into the upper level of the laser transition. Due to the fast decay, the level 3 may be assumed to be nearly unoccupied. Therefore, for each atom, we have

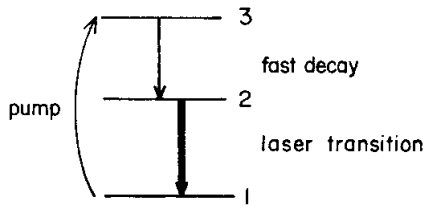


FIG. 1

$$(N_1 + N_2)_\mu = 1 \tag{16}$$

Summing up the eqns (14), (15) over all atoms and using (8) and (16), we find

$$\frac{dD}{dt} = \frac{D_0 - D}{T} - 2DWn \tag{17}$$

where

$$D_0 = \frac{w_{12} - w_{21}}{w_{12} + w_{21}} N, \qquad \frac{1}{T} = w_{12} + w_{21}, \tag{18}$$

and N is the total number of atoms. The *unsaturated inversion*, D_0, is obtained by the pump and all other *incoherent* processes but in the absence of laser action (i.e. $n = 0$). The actual, or saturated inversion is D. Equations (9) and (17) are the complete laser equations.

2.3. STATIONARY SOLUTION OF EQUATIONS (9) AND (17)

When the system is stationary, we have

$$dn/dt = 0 \quad \text{and} \quad dD/dt = 0. \tag{19}$$

From (17) we obtain

$$D = D_0/(1+2TWn), \tag{20}$$

which determines the saturated inversion D by means of the unsaturated inversion D_0 and the photon number n. Inserting (20) into (9) and solving the resulting equation for n we obtain

$$n = (D_0 W - 2\kappa)/4TW\kappa, \tag{21}$$

where we have neglected the spontaneous emission term. This equation allows us to determine the actual photon number as a function of the unsaturated inversion. When we require that the number n starts to be positive, we reobtain the threshold condition (10). There is a strange difficulty with rate equations. The photon number ought to be positive even if (10) is not fulfilled. We might try to include the spontaneous emission term (1), but then we mix two processes which, intuitively, apply to different situations of "coherence". This problem has been solved in a satisfactory way only recently; its solution will be discussed in Section 4.

2.4. RATE EQUATIONS FOR THE THREE-LEVEL SYSTEM

In most cases laser action occurs between two excited levels (see Fig 2.). The field equation remains the same (see eqn (9)), but the matter equations become

$$\mathrm{d}N_{2,\mu}/\mathrm{d}t = -w_{21} N_{2,\mu} + w_{02} N_{0,\mu} - W(N_2 - N_1)_\mu n, \tag{22}$$

$$\mathrm{d}N_{1,\mu}/\mathrm{d}t = -w_{10} N_{1,\mu} + w_{21} N_{2,\mu} + w_{01} N_{0,\mu} + W(N_2 - N_1)_\mu n, \tag{23}$$

$$\mathrm{d}N_{0,\mu}/\mathrm{d}t = -w_{02} N_{0,\mu} + w_{10} N_{1,\mu}. \tag{24}$$

We shall discuss some important special cases of (22–24).

Because the electron is in any of the three levels, we have

$$N_{0,\mu} + N_{1,\mu} + N_{2,\mu} = 1, \tag{25}$$

which allows $N_{0,\mu}$ to be eliminated. In many cases the ground state may be

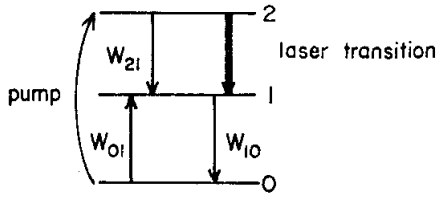


FIG. 2

treated as a reservoir. If the pump rates w_{02}, w_{01} are much slower than the decay rates w_{21} and w_{10}, most of the atoms are in their ground states, so that

$$N_{0,\mu} \approx 1. \tag{26}$$

Using (26) in eqns (22) and (23) we obtain

$$dN_{2,\mu}/dt = w_{02} - w_{21} N_{2,\mu} - W(N_2 - N_1)_\mu n \tag{27}$$

$$dN_{1,\mu}/dt = w_{01} - w_{10} N_{1,\mu} + W(N_2 - N_1)_\mu n. \tag{28}$$

In the second equation we have also neglected a term $w_{21}N_{2,\mu}$ in order to be in accordance with other approaches. Without laser action, $N_{2,\mu}$ and $N_{1,\mu}$ would arrive independently at their equilibrium values (w_{02}/w_{21}) and (w_{01}/w_{10}), respectively.

The laser equation can be simplified further if the recombination rate w_{10} is high compared to all other rates. Then we have

$$N_{1,\mu} \approx 0 \quad \text{and} \quad D \approx \sum_{\mu=1}^{N} N_{2,\mu}. \tag{29}$$

In this case we may drop eqn (28). Summing eqn (27) over μ we find

$$dN_{2,\text{tot}}/dt = Nw_{02} - w_{21} N_{2,\text{tot}} - WN_{2,\text{tot}} n. \tag{30}$$

This equation allows us to transcribe all results obtained for the two-level system to the three-level system; $N_{2,\text{tot}}$ replaces D, with the only difference that $2W$ in eqn (17) is replaced by W in eqn (30).

2.5. TIME-DEPENDENT SOLUTIONS:[4] RELAXATION OSCILLATIONS WITH SMALL AMPLITUDES[5]

In contrast to the stationary case, where we can find the exact solution, the time dependent case is much more complicated because the eqns (9) and (30) are nonlinear. However, for small oscillations around the equilibrium values, these equations can be linearized. We treat the upper laser transition, and start from the eqns (9) and (30). We put

$$D \approx N_{2,\text{tot}} = N^0_{2,\text{tot}} + \delta N_{2,\text{tot}} \tag{31}$$

and

$$n = n_0 + \delta n, \tag{32}$$

where the stationary solutions n_0 and $N^0_{2,\text{tot}}$ are fixed by the conditions

$$-2\kappa + WN^0_{2,\text{tot}} = 0 \tag{33}$$

and

$$Nw_{02} - N^0_{2,\text{tot}} w_{21} - n_0 N^0_{2,\text{tot}} W = 0 \tag{34}$$

Inserting (31) and (32) into (9) and (30) using (33) and (34) and linearizing we obtain, after a slight rearrangement, the following equations

$$\frac{\mathrm{d}}{\mathrm{d}t}\delta n = \frac{\delta N_{2,\mathrm{tot}}}{N^0_{2,\mathrm{tot}}} 2\kappa n_0 \tag{35}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\delta N_{2,\mathrm{tot}} = -\frac{\delta N_{2,\mathrm{tot}}}{N^0_{2,\mathrm{tot}}} N w_{02} - \delta n N^0_{2,\mathrm{tot}} W. \tag{36}$$

In order to solve this system of linear homogeneous differential equations with constant coefficients we put

$$\delta n = A \exp(\alpha t), \tag{37}$$

$$\delta N_{2,\mathrm{tot}} = B \exp(\alpha t), \tag{38}$$

and α has then to obey the equation

$$\alpha^2 + (w_{02} N W/2\kappa)\alpha + (W N w_{02} - 2w_{21}\kappa) = 0. \tag{39}$$

According to the two roots of eqn (39), δn can be written in the form

$$\delta n = A_1 \exp[-(\Gamma - i\omega_r)t] + A_2 \exp[-(\Gamma + i\omega_r)t], \tag{40}$$

where

$$\Gamma = w_{02} w_{21}/2w_{\mathrm{thr}} \tag{41}$$

and ω_r and ω_{thr}, respectively, are given by

$$\omega_r = \left[-\frac{w^2_{02}\, w^2_{21}}{4w^2_{\mathrm{thr}}} + \left(\frac{w_{02}}{w_{\mathrm{thr}}} - 1\right) 2\kappa w_{21} \right]^{\frac{1}{2}} \tag{42}$$

$$w_{\mathrm{thr}} = 2w_{21}\kappa/NW. \tag{43}$$

It is evident from (40) that the photon number, after a deviation δn from its equilibrium, returns to its equilibrium value n_0. The relaxation oscillations occur with the frequency ω_r.

2.6. THE GIANT PULSE LASER

Another example for the application of rate equations is the giant pulse laser. It should be noted, however, that phase-locking effects may play a very important role in the formation of short pulses, and since phase information is not included in the rate equations, they will not predict these effects.

Let us consider a resonator whose quality factor Q is kept very low at the beginning so that no laser action can occur. Then the pumping drives the popu-

ulation inversion to a higher value than that which can be maintained during the laser process. If now the Q is suddenly increased, the feedback causes laser action at a considerably higher output power level than can be achieved in steady-state operation. This method, called Q-spoiling,[6] can be experimentally verified by rotating mirrors (or prisma, or shutter discs)[7] so that the reflectivity of one mirror changes periodically with time. The switching can also be achieved by a Kerr cell,[8] which is particularly suitable, if the light of the laser-active atoms is already polarized (especially of ruby crystals, for example).

Instead of changing the loss, we can change the gain. For example, in ruby, we can cause a quasi-Stark effect, which splits the atomic line, thus giving rise to an increased effective linewidth and a lowered gain. When the electric field is released, the laser pulse builds up.[9]

The whole time-evolution of this process can be treated by means of the eqns (9) and (30) which we solve semi-quantitatively. In the beginning ($t = 0$) the Q-value is kept low (i.e. $2\kappa_0$ is kept high). Under a steady pumping D and n assume steady state values D_i, n_i. Note that D_i is essentially determined by *spontaneous emission.* At $t=0$ we switch the resonator suddenly to a high Q (low $2\kappa_1$). Because the losses are low, we expect the laser pulse to build up so quickly by induced emission that the spontaneous emission rate (as well as nonradiative decay rates) can be neglected. Furthermore we neglect the further pumping. The laser equations then take the form

$$\mathrm{d}n/\mathrm{d}t = (WD - 2\kappa_1)\, n, \tag{44}$$

$$\mathrm{d}D/\mathrm{d}t = -2WnD. \tag{45}$$

According to (45), the inversion D decreases rather slowly for small values of n, so that we may replace D on the right hand sides of (44), (45) by its initial value D_i. The solutions of (44) and (45) then read

$$n = n_i \exp(\alpha t), \tag{46}$$

$$D = D_i \{1 + 2Wn_i[1 - \exp(\alpha t)]/\alpha\}, \tag{47}$$

$$\alpha = WD_i - 2\kappa_1.$$

In the first phase of the pulse, n increases exponentially with the gain constant α and then, because of (45) or (47), the inversion decreases. The increase stops certainly at a time t_1, when $D(t_1) \approx 0$ (in fact, it stops earlier due to the loss-term $2\kappa_1$). Neglecting 1 compared to $\exp(\alpha t)$, it follows from (47) that

$$n_i \exp(\alpha t_1) = \alpha/2W = n_{\max} \tag{48}$$

or

$$t_1 = (1/\alpha) \ln(\alpha/2Wn_i). \tag{49}$$

Equation (48) represents incidentally the number of photons present at this time, t_1. After the time t_1, we may assume $D \approx 0$. The photon number then decays approximately exponentially

$$n(t) = n_{\max} \exp\left[-2\kappa_1(t-t_1)\right]. \tag{50}$$

The equations we have presented are at least good enough for rough estimates of the quantities t_1, $n_{\max}$ and $n(t)$. For a more detailed analysis, the eqns (44) and (45) must be solved in the vicinity of $t = t_1$ by a computer (the slopes are well represented by (46) and (50)). This has been performed by, for example, Wagner and Lengyel,[10] who have also obtained several exact relations.

2.7. GENERAL FORMULATION

Above we have treated the example of a single mode interacting with a set of two- or three-level atoms. We now want to formulate the rate equations quite generally for M modes and a system of L-level atoms. In particular we shall take spatial effects into account. The modes are distinguished by an index λ, where λ not only refers to the wavelength, but also to all other spatial properties of the modes. These cavity modes interact with N atoms at random lattice sites, x_μ. The optical transition shall occur between the levels 2 and 1. The levels 1 and 2 may have any position in the system. The other levels participate in the pumping process. Again the rate equations can be divided into two groups, (α) the field equations, and (β) the matter equations.

(α) *The field equations* describe the rate of change of the mean photon number n_λ of the mode λ under the influence of losses and gain. The losses can be caused by transmission through the end-faces, diffraction, scattering by impurity centres, crystal inhomogeneities, etc. These effects are taken care of by attributing a decay time $t_\lambda = \kappa_\lambda/2$ to the mode λ. The loss rate is thus given by

$$-2\kappa_\lambda n_\lambda. \tag{51}$$

The gain is due to spontaneous and induced emission. The spontaneous decay rate from the level 2 of the atom μ into the mode λ is proportional to the occupation number $N_{2,\mu}$ of the upper laser-active state 2 of the atom μ. We may further guess, that it is proportional to the mode intensity at the atomic position x_μ (at least in the dipole approximation).

If $U_\lambda(x_\mu)$ is the amplitude of mode λ, then its relation with the optical transition rate is

$$W \propto |U_\lambda(x_\mu)|^2. \tag{52}$$

A typical example is $U_\lambda(x_\mu) = \sqrt{(2/V)} \sin k_\lambda x_\mu$ where V is the volume of the cavity. Furthermore if the line connected with the optical transition $2 \to 1$ has

a Lorentzian shape of half-width γ, we expect that in the case of detuning, we have

$$W \propto \frac{2\gamma}{\gamma^2 + (\nu_\mu - \omega_\lambda)^2}, \tag{53}$$

where ν_μ is the (circular) centre frequency of atom μ and ω_λ is the frequency of the mode λ in the unloaded cavity. Finally, we expect that W goes with the square of the atomic transition dipole moment θ_{21}, taken in the direction of the mode polarization $\mathbf{e}_\lambda$, i.e.

$$W \propto |\mathbf{e}_\lambda \boldsymbol{\theta}_{21}|^2. \tag{54}$$

These considerations make it clear why the exact derivation of W yields

$$W^0_{\lambda,\mu} = \frac{2\gamma}{\gamma^2 + (\nu_\mu - \omega_\lambda)^2} |g_{\mu\lambda}|^2, \tag{55}$$

where

$$g_{\mu\lambda} = i\boldsymbol{\theta}_{21} \left(\frac{2\pi\omega_\lambda}{\hbar}\right)^{\frac{1}{2}} \mathbf{U}_\lambda(\mathbf{x}_\mu). \tag{56}$$

(or $g_{\mu\lambda} = g\sqrt{(V)}\, U_\lambda(x_\mu)$, where g does not depend on λ, if we restrict our analysis to a single polarization). We have supplied W with all necessary indices: 0 means optical, λ refers to the mode, μ to the atom.

The induced emission and absorption rates can be obtained in complete analogy to Section 2.1. The net gain is given by

$$n_\lambda W^0_{\lambda\mu}(N_2 - N_1)_\mu. \tag{57}$$

We thus obtain the *field equations*

$$\dot{n}_\lambda = -2\kappa_\lambda n_\lambda + n_\lambda \sum_\mu W^0_{\lambda\mu}(N_2 - N_1)_\mu + \sum_\mu W^0_{\lambda\mu} N_{2,\mu}. \tag{58}$$

(β) *The matter equations* can be derived quite similarly. For the levels participating in laser process we obtain

$$\dot{N}_{2,\mu} = \sum_{l\neq 2} w_{l,2} N_{l,2} - N_{2,\mu} \sum_{l\neq 2} w_{2,l} - \sum_\lambda n_\lambda W^0_{\lambda\mu}(N_2 - N_1)_\mu. \tag{59}$$

$$\dot{N}_{1,\mu} = \sum_{l\neq 1} w_{l,1} N_{l,\mu} - N_{1,\mu} \sum_{l\neq 1} w_{1,l} + \sum_\lambda n_\lambda W^0_{\lambda\mu}(N_2 - N_1)_\mu. \tag{60}$$

For the other levels which are used by the pumping process we get

$$\dot{N}_{m,\mu} = \sum_{l\neq m} w_{lm} N_{l,\mu} - N_{m,\mu} \sum_{l\neq m} w_{ml}. \tag{61}$$

The eqns (59)–(61) are not independent of each other. If we sum up these equations over all levels of the atom μ, we obtain

$$(\mathrm{d}/\mathrm{d}t)\left(\sum_l N_{l,\mu}\right) = 0, \tag{62}$$

so that

$$\sum_l N_{l,\mu} = \text{constant}. \tag{63}$$

The fact that the sum over the occupation numbers, $N_{l,\mu}$, of a single atom is a time independent constant is a consequence of the conservation of the electron number in a single atom. Hence we have for the single electron problem under consideration

$$\sum_l N_{l,\mu} = 1. \tag{64}$$

The transition rate from level l to level m is w_{lm}, and is caused by external pumplight, collisions of the second kind, excitation collisions with electrons, etc., and all nonradiative and radiative spontaneous transitions. In the following we drop the term $\sum_\mu W^0_{\lambda\mu} N_{2,\mu}$ in eqn (58) since this expression describes the generation of "incoherent photons". (For a deeper motivation and a detailed discussion see Section 4.)

2.8. TREATMENT OF THE STEADY STATE

The main task consists in the determination of the photon number as a function of the pumping, and especially in the derivation of a threshold condition. We describe the general procedure. In the steady state we put

$$\dot{n}_\lambda = \dot{N}_{l,\mu} = 0. \tag{65}$$

From (59), (60) and (61) we obtain for the inversion $d_\mu = (N_2 - N_1)_\mu$ an expression of the form

$$d_\mu = \frac{A_\mu}{B_\mu + (n_1 W^0_{1,\mu} + n_2 W^0_{2,\mu} + \ldots + n_M W^0_{M,\mu})\, C_\mu}. \tag{66}$$

A_μ, B_μ, C_μ do not depend on the n_λ's (but still contain the transition rates, w_{lm}). Equation (66) can be derived as follows. Because of (64) we need only consider eqns (59), (61). We drop, for example, the equations with index 1 and introduce the new variables $(N_2 - N_1)$ and $(N_2 + N_1)$ which replace the variables N_2 and N_1. The n_λ's then appear in only a single equation where they act as factors of $d_\mu = (N_2 - N_1)_\mu$ in the form

$$\sum_\lambda n_\lambda W^0_{\lambda,\mu}. \tag{67}$$

Using the general theorem about the solution of linear equations by means of determinants we can write

$$d_\mu = \Delta_1/\Delta \tag{68}$$

where the total determinant Δ depends on (67) at most in a linear fashion, because (67) occurs only in a single element of the entire Δ. Furthermore, Δ_1 is obtained from Δ in such a way that the column containing (67) is cancelled and replaced by the inhomogeneity introduced by eqn (64). Consequently, Δ_1 does not depend on the n_λ's. For an explicit evaluation of the A's B's and C's it is advisable to treat the 3- and 4-level system explicitly. The *photon numbers* (and thus the *emitted coherent power*) can be determined as follows. We assume that M modes show laser action so that $n_\lambda > 0$ for $\lambda = 1, \ldots, M$ (λ', λ'' also denote modes) After dividing the eqn (58) by n_λ, we find

$$2\kappa_\lambda = \sum_\mu W^0_{\lambda,\mu}(N_2 - N_1)_\mu \tag{69}$$

or, on account of (66),

$$2\kappa_\lambda = \sum_\mu \frac{W^0{}_{\lambda\mu} A_\mu}{B_\mu + C_\mu \sum_{\lambda'} n_{\lambda'} W^0_{\lambda',\mu}}. \tag{70}$$

There are M equations for the M unknowns, n_λ, for which the condition $n_\lambda > 0$ holds. If it turns out that some $n_{\lambda''} \leqslant 0$, these $n_{\lambda''}$'s *and* the corresponding eqn (70) must be dropped. In order to obtain an explicit solution of eqn (70) it is generally necessary to treat special cases.

2.9. A THEOREM ON THE MODE SELECTION IN THE COMPLETELY HOMOGENEOUS CASE

We assume that the atomic line is *homogeneously* broadened so that the transition frequencies ν_μ are equal ($=\nu_0$). We represent the modes by running waves so that they possess a spatially independent intensity. Then the optical transition rate does not depend on μ. Finally, it is assumed that the pumping process (and the decay rates) are spatially homogeneous. Under these assumptions, A, B, C do not depend on the atomic index μ so that (70) simplifies to

$$\sum_{\lambda'} n_{\lambda'} W_{\lambda'}{}^0 = \frac{N W_\lambda{}^0 A}{2\kappa_\lambda C} - \frac{B}{C}. \tag{71}$$

Because the left-hand side does not depend on the mode-index λ, we find that

$$W_\lambda{}^0/\kappa_\lambda. \tag{72}$$

must be *independent of* λ. Consequently, the eqn (71) can be solved in a consistent way only for those modes for which the condition (72) is fulfilled.

From (71) it can be deduced that the mode for which $W_\lambda{}^0$ has a maximum, starts laser action first. If this maximum is assumed only for one mode, then this mode remains the only laser mode for every pump strength. *In the completely homogeneous case only a single mode (or degenerate modes in the sense of* (72) *with maximum* (W^0/κ)*) can oscillate.*[11]

The *laser condition* is obtained from (69) by putting all n_λ's equal zero. Using (55) we find

$$2\kappa = (N_2 - N_1)_{\text{threshold}} \cdot N \frac{2\gamma|g|^2}{\gamma^2 + (\omega - \nu_0)^2}. \tag{73}$$

2.10. THE COEXISTENCE OF MODES CAUSED BY SPATIAL INHOMOGENEITIES IN A HOMOGENEOUSLY BROADENED LASER

We consider the case in which the line is homogeneously broadened and the modes depend on the space points, for instance, like a standing wave. We shall base our treatment of the problem on the following *approximation for the determination of the photon-numbers*. We have seen above the photon numbers, n_λ, can be obtained in principle" by solving the eqn (70). This requires, however, that the sums over μ are performed, but these can be done explicitly only in very special cases. We therefore expand the right-hand side of eqn (70) into a power series with respect to $\sum_{\lambda'} n_{\lambda'} W_\lambda{}^0$, where we usually confine ourselves to terms linear in the photon number. As long as the laser does not operate too highly above threshold, the photon numbers are still small, so that this approximation is well justified and can be substantiated by numerical examples. We thus obtain from (58) the following set of linear inhomogeneous equations for n_λ (with the supplementary condition $n_\lambda > 0$),

$$\sum_{\mu,\lambda'} \frac{A_\mu C_\mu}{B_\mu{}^2} W^0_{\lambda,\mu} W^0_{\lambda',\mu} n_{\lambda'} = -2\kappa_\lambda + \sum_\mu W^0_{\lambda,\mu} \frac{A_\mu}{B_\mu}. \tag{74}$$

From (74) again the threshold condition follows by putting all n_λ's equal zero.

As an explicit example,[12] we treat modes whose space dependence is given by $\sin k_\lambda x_\mu$ so that

$$W^0_{\lambda,\mu} \sim \sin^2 k_\lambda x_\mu. \tag{75}$$

We shall consider single mode operation first. The expressions for d_μ show that the inversion is unaltered where the mode has its node. Thus a second mode with a different wavelength can still "eat" from those portions of the crystal in which the inversion is not appreciably diminished by the first mode. We

shall confine ourselves to the one-dimensional case, as indicated by (75), or to modes with **k**-vectors which differ only in one component. We then obtain

$$\sum_{\mu} W^0_{\lambda,\mu} = W_\lambda,$$

$$N\sum_{\mu} W^0_{\lambda,\mu} W^0_{\lambda',\mu} = \tfrac{3}{2} W_\lambda^2, \quad \text{for} \quad \lambda = \lambda', \tag{76}$$

$$= W_\lambda W_{\lambda'}, \quad \text{for} \quad \lambda \neq \lambda',$$

where $$W_\lambda = 2g^2\gamma/\{(\nu_0-\omega_\lambda)^2+\gamma^2\} N.$$

The system of eqn (74) thus takes the form

$$3W_\lambda n_\lambda + 2\sum_{\lambda'\neq\lambda} W_{\lambda'} n_{\lambda'} = 2\frac{B}{C}\left(1-\frac{2\kappa_\lambda}{d_0 W_\lambda}\right) N = l_\lambda, \tag{77}$$

where we have put $A/B = d_0$. Introducing $W_\lambda n_\lambda$ as new variables, we observe that (77) has a completely symmetric form, which immediately allows the explicit solution

$$W_\lambda n_\lambda = l_\lambda - \frac{2(l_1+\ldots+l_m)}{2M+1}$$

$$= \frac{2BN}{C}\left(\frac{1}{2M+1} - \frac{2\kappa_\lambda}{d_0 W_\lambda} + \frac{4\{(\kappa_1/W_1)+\ldots+(\kappa_m/W_m)\}}{(2M+1)\,d_0}\right), \tag{78}$$

where M is the number of coexisting modes.

For the further evaluation of eqn (78) we treat two examples, α and β, below.

(α) Axial modes with a different frequency distance from the line centre

Let the axial modes be discriminated from the nonaxial modes by a large difference between their losses so that we can confine ourselves to the axial modes alone. These modes shall have the same losses, κ, but they are still discriminated by their different frequencies, ω_m. Putting $\omega_\lambda = \nu_0 + m_\lambda\delta$ where ν_0 is the frequency of the atomic line centre, m_λ is the mode index and δ is the frequency spacing, we obtain the following as the condition for $n_\lambda > 0$ (where $\omega_\lambda = \nu_0 + m_0\delta$)

$$\frac{1}{2M+1} - \frac{\kappa}{\gamma g^2 d_0}(\gamma^2 + m_0{}^2\delta^2) + \frac{1}{2M+1}\left(\frac{2\kappa\gamma M}{d_0 g^2} + \frac{2\kappa\delta^2}{d_0\gamma g^2}\sum m^2\right) > 0, \tag{79}$$

or

$$1 - \frac{\kappa\gamma^2}{\gamma g^2 d_0} - \frac{\kappa\delta^2}{\gamma g^2 d_0}\{m_0{}^2(2M+1) - 2\sum m^2\} > 0. \tag{80}$$

Because we have assumed a symmetric position of the mode frequencies with respect to the atomic line centre, we have

$$m_0 = (M-1)/2, \tag{81}$$

where m_0 is the index of the mode with maximum separation from line centre. Thus we can write eqn. (80) finally in the form

$$1-\frac{\kappa\gamma^2}{\gamma g^2 d_0}-\frac{\kappa\delta^2}{\gamma g^2 d_0}\{m_0{}^2(4m_0+3)-\tfrac{2}{3}m_0(m_0+1)(2m_0+1)\}>0. \tag{82}$$

The higher the inversion d_0 and the smaller the frequency difference (compared to the linewidth γ), the bigger is m_0, and thus, the number, M, of coexisting modes.

(β) Axial modes with different losses

We assume that the mode with the lowest κ lies exactly at resonance, and that the other modes which are lying close to the resonance are non-axial modes. For the sake of simplicity we assume that all these modes are situated in the same plane in **k**-space. Because the spatial modulation varies now in a single direction (perpendicular to the laser axis), a spatially inhomogeneous inversion results, so that again a factor 3/2 occurs in eqn (76). (The modes shall have radial symmetry). We can neglect the change in frequency but we have to take into account the fact that the losses, κ_λ, increase with an increasing angle between the propagation direction of the modes and the laser axis. We put, therefore,

$$\kappa_m = \kappa(1+q)^{m-1}. \tag{83}$$

where q is a small dimensionless quantity.
Inserting (83) into eqn (78) yields an expression for the number of modes, M, which is

$$M^2 < \frac{d_0 W}{2\kappa q}\left(1-\frac{2\kappa}{d_0 W}\right). \tag{84}$$

When we express the inversion d_0 by the threshold inversion $d_c = 2\kappa/W$ in the form $d_0 = d_c(1+\eta)$ eqn (84) can be further simplified to give

$$M^2 \leqslant \eta/q. \tag{85}$$

If, for instance, $q = 10^{-2}$, i.e. if the losses differ only by 1 %, the coexistence of modes becomes possible by increasing the inversion by a few percent. The coexistence of modes can be weakened by cross-relaxation between atoms at different space-points or by diffusion processes so that the spatial dependence of the inversion is flattened.

3. The Semiclassical Approach[13]

Heuristic derivation of the basic equations

In the semiclassical approach, the electro-magnetic field is treated classically by means of Maxwell's equations, but matter (i.e. the electrons) is treated quantum-mechanically. To connect both treatments properly, it is necessary to describe the dynamics of the electronic motion by equations referring to *averages* which are quantum-mechanical and statistical in nature. Because this approach can be fully understood only in a completely quantum mechanical treatment of the laser, we postpone the rigorous derivation to Section 4 and confine our present treatment of the semiclassical equations to a *heuristic* derivation. We start with the wave-picture and derive the mode-picture afterwards.

3.1. Wave-picture[14]

a. The field equations

From Maxwell's equation

$$\operatorname{curl} \mathbf{H} = 4\pi \mathbf{j}/c + (\dot{\mathbf{E}} + 4\pi \dot{\mathbf{P}})/c, \tag{86}$$

and using the relations†

$$\mathbf{j} = \sigma \mathbf{E}, \quad \mathbf{H} = \operatorname{curl} \mathbf{A}, \quad \mathbf{E} = -\dot{\mathbf{A}}/c \tag{87}$$

and the Coulomb gauge

$$\operatorname{div} \mathbf{A} = 0, \tag{88}$$

we obtain a differential equation for the vector potential $\mathbf{A}$

$$-\Delta \mathbf{A} + \frac{1}{c^2} \ddot{\mathbf{A}} + \frac{4\pi\sigma}{c^2} \dot{\mathbf{A}} = \frac{4\pi}{c} \dot{\mathbf{P}} \tag{89}$$†

and, by differentiation with respect to the time, a differential equation for the electric field strength

$$-\Delta \mathbf{E} + \frac{1}{c^2} \ddot{\mathbf{E}} + \frac{4\pi\sigma}{c^2} \dot{\mathbf{E}} = -\frac{4\pi}{c^2} \ddot{\mathbf{P}} \tag{90}$$

b. The material equations

The polarization $\mathbf{P}$ in eqn (90) is not a fixed source, but is caused by the electric field, $\mathbf{E}$. We adopt in this chapter the following laser-model. The material consists of a solid state matrix, e.g. a crystalline lattice, in which the

† note, that σ may be a tensor.

laser-active atoms labelled with an index μ, are embedded at random lattice sites $\mathbf{x}_\mu$. We assume that the solid state matrix causes a polarization, which is proportional to $\mathbf{E}$. The effect of the inactive solid state matrix may be taken care of by using an effective light velocity. In the following we therefore consider only that part of $\mathbf{P}$ which stems from the laser-active atoms. We decompose $\mathbf{P}$ into the single atom contributions

$$\mathbf{P}(\mathbf{x}) = \sum_\mu \delta(\mathbf{x}-\mathbf{x}_\mu)\,\mathbf{p}_\mu, \tag{91}$$

where δ is Dirac's δ-function and the sum runs over the laser active atoms. Each $\mathbf{p}_\mu$ can be visualized as decomposed into $e\mathbf{x}$ where e is the electronic charge and $\mathbf{x}$ is the displacement of a local oscillator. In a *purely classical* treatment, the equation for a component of $\mathbf{p}_\mu$ would read (as in the usual dispersion theory),

$$\ddot{\mathbf{p}}_\mu + 2\gamma\,\dot{\mathbf{p}}_\mu + \nu_\mu^2\,\mathbf{p}_\mu = \frac{e^2}{m}\,\mathbf{E}(\mathbf{x}_\mu), \tag{92}$$

where ν_μ is the frequency of atomic polarization. The damping constant γ can be identified with the atomic width. This equation is, however, insufficient, because it does not account for the effect of saturation. The quantum mechanical calculation shows that the strength and the sign (i.e. the relative phase) between $\mathbf{p}$ and the driving field depends on the atomic inversion, which is defined as the difference $d_\mu = (N_2 - N_1)_\mu$ between the occupation numbers of the upper and lower level of the lasing atom, μ. Anticipating further a quantum mechanical replacement of the factor (e^2/m), we may write the quantum mechanical analogue of (92) in the form

$$\ddot{\mathbf{p}}_\mu + 2\gamma\,\dot{\mathbf{p}}_\mu + \nu_\mu^2\,\mathbf{p}_\mu^2 = -\,2\omega_0\,\frac{|\boldsymbol{\theta}_{12}^2|}{\hbar}\,\mathbf{E}(\mathbf{x}_\mu)\,d_\mu, \tag{93}$$

where $\boldsymbol{\theta}_{12}$ is the atomic dipole moment matrix element and ω_0 is the average mode frequency. For simplicity we have assumed that the vectors $\boldsymbol{\theta}_{12}$, $\mathbf{p}_\mu$ and $\mathbf{E}$ are parallel, so that $\boldsymbol{\theta}_{12}$ enters eqn (93) in the given way.

Multiplying eqn (93) by $\delta(\mathbf{x}-\mathbf{x}_\mu)$ and summing over μ we obtain (provided the atomic line is homogeneously broadened, i.e. $\nu_\mu \equiv \nu$)

$$\ddot{\mathbf{P}}(\mathbf{x},t) + 2\gamma\,\dot{\mathbf{P}}(\mathbf{x},t) + \nu^2\,\mathbf{P}(\mathbf{x},t) = 2\omega_0\,\frac{|\boldsymbol{\theta}_{12}^2|}{\hbar}\,\mathbf{E}(\mathbf{x})\,d(\mathbf{x}), \tag{94}$$

where $d(\mathbf{x})$ is defined by

$$d(\mathbf{x}) = \sum_\mu \delta(\mathbf{x}-\mathbf{x}_\mu)\,d_\mu, \tag{95}$$

The inversion (population difference) $d_\mu = (N_2 - N_1)_\mu$ itself may change due to radiative emission and due to relaxation processes (which include the pumping of the atom). Accordingly, the rate of change of the atomic energy $\hbar\nu d_\mu$ is given by

$$\frac{\mathrm{d}}{\mathrm{d}t}(\hbar\nu d_\mu) = \left(\frac{\mathrm{d}(\hbar\nu d_\mu)}{\mathrm{d}t}\right)_{\text{field}} + \left(\frac{\mathrm{d}(\hbar\nu d_\mu)}{\mathrm{d}t}\right)_{\text{relaxation}}, \tag{96}$$

where

$$\left(\frac{\mathrm{d}(\hbar\nu d_\mu)}{\mathrm{d}t}\right)_{\text{field}} = 2\mathbf{E}(\mathbf{x}_\mu)\cdot\dot{\mathbf{p}}_\mu, \tag{97}$$

is the usual classical expression.

If we confine ourselves *to a two level atom* we have

$$\left(\frac{\mathrm{d}(\hbar\nu d_\mu)}{\mathrm{d}t}\right)_{\text{relaxation}} = \frac{\hbar\nu\,(d_0 - d)_\mu}{T}, \tag{98}$$

where T is the effective relaxation time to restore the equilibrium value d_0.

Putting (96), (97) and (98) together we find

$$\frac{\mathrm{d}}{\mathrm{d}t}d_\mu = \frac{(d_0 - d)_\mu}{T} + \frac{2}{\hbar\nu}\mathbf{E}(\mathbf{x}_\mu)\cdot\dot{\mathbf{p}}_\mu. \tag{99}$$

Multiplying eqn (99) with $\delta(\mathbf{x} - \mathbf{x}_\mu)$ and summing over μ we obtain for the inversion density $d(\mathbf{x})$

$$\frac{\mathrm{d}}{\mathrm{d}t}d(\mathbf{x}) = \frac{(d_0 - d)(\mathbf{x})}{T} + \frac{2}{\hbar\nu}\mathbf{E}(\mathbf{x})\cdot\dot{\mathbf{P}}(\mathbf{x}). \tag{100}$$

Equations (90), (93) and (99) are the basic equations for a set of laser modes interacting with a set of two-level atoms. In the case of multi-level atoms the eqns (100) and (93) remain valid, but (99) acquires a more complicated structure. The eqns (90), (94) and (100) describe the same problem in terms of a continuous polarization and a continuous inversion density. These equations may be used as a starting point for the treatment of pulse propagation in laser-active media, like π-pulses, 2π-pulses (i.e. self-induced transparency), and self-pulsing.[14] For a treatment of some of these effects see the chapter by Bloembergen in this volume. In the following, we treat explicitly eqns (90), (93) and (99) and leave it to the reader to perform the same steps with eqns (90), (94) and (100).

3.2. MODE PICTURE

We decompose $\mathbf{E}(\mathbf{x}, t)$ into the normalized eigenfunctions $\mathbf{U}_\lambda(\mathbf{x})$ of the unloaded interferometer ("cavity")

$$\mathbf{E}(\mathbf{x}, t) = \sum_\lambda E_\lambda(t)\,\mathbf{U}_\lambda(\mathbf{x}). \tag{101}$$

The $\mathbf{U}$'s obey the homogeneous wave equation, so that

$$\Delta\mathbf{U}_\lambda = -k_\lambda{}^2\,\mathbf{U}_\lambda = -\frac{\omega_\lambda{}^2}{c^2}\,\mathbf{U}_\lambda. \tag{102}$$

Because the $\mathbf{U}$'s are defined within an open cavity, they are not exactly orthogonal, although for practical purposes it is sufficient to assume their orthogonality. The open cavity causes diffraction losses, which may be taken care of by an "effective" conductivity σ in eqn (90). Inserting (101) into (90), multiplying the resulting equation with $\mathbf{U}_\lambda(\mathbf{x})$ and integrating over the volume of the "cavity" we find

$$\omega_\lambda{}^2 E_\lambda + \ddot{E}_\lambda + \sum_{\lambda'} 4\pi\sigma_{\lambda\lambda'}\dot{E}_{\lambda'} = -4\pi\ddot{P}_\lambda, \tag{103}$$

where

$$\sigma_{\lambda\lambda'} = \int \mathbf{U}_\lambda \sigma \mathbf{U}_{\lambda'}\,\mathrm{d}V \tag{104}$$

and

$$\ddot{P}_\lambda = \int \mathbf{U}_\lambda\,\ddot{\mathbf{P}}\,\mathrm{d}V. \tag{105}$$

We abbreviate $2\pi\sigma_{\lambda\lambda} = \kappa_\lambda$, where κ_λ is the cavity halfwidth of mode λ and $2\pi\sigma_{\lambda\lambda'} = \kappa_{\lambda\lambda'}$, $(\lambda \neq \lambda')$. In the following we put $\sigma_{\lambda\lambda'} = 0$ for $\lambda \neq \lambda'$.

The matter equations can be immediately found from eqns (93) and (99) by inserting the expansion (101) for $\mathbf{E}$, which yields

$$\ddot{\mathbf{p}}_\mu + 2\gamma\,\dot{\mathbf{p}}_\mu + \nu_\mu{}^2\,\mathbf{p}_\mu = -2\omega_0\,\frac{|\boldsymbol{\theta}_{12}^2|}{\hbar}\,d_\mu \sum_\lambda E_\lambda(t)\,\mathbf{U}_\lambda(\mathbf{x}_\mu), \tag{106}$$

and

$$\dot{d}_\mu = \frac{d_0 - d_\mu}{T} + \frac{2}{\hbar\nu_\mu}\,\dot{\mathbf{p}}_\mu \sum_\lambda E_\lambda(t)\,\mathbf{U}_\lambda(\mathbf{x}_\mu). \tag{107}$$

3.3. THE ROTATING WAVE APPROXIMATION

This approximation can be applied to both the wave and mode picture. We shall treat explicitly the mode picture and decompose E_λ and $\mathbf{p}_\mu$ into a positive and negative frequency part to obtain

$$E_\lambda(t) = \exp(-i\omega_\lambda t)\,\tilde{E}_\lambda{}^{(+)}(t) + \exp(i\omega_\lambda t)\,\tilde{E}_\lambda{}^{(-)}(t) \equiv E_\lambda{}^{(+)}(t) + E_\lambda{}^{(-)}(t), \tag{108}$$

$$\mathbf{p}_\mu(t) = \exp(-i\nu_\mu t)\,\tilde{\mathbf{p}}_\mu{}^{(+)}(t) + \exp(i\nu_\mu t)\,\tilde{\mathbf{p}}_\mu{}^{(-)}(t) \equiv \mathbf{p}_\mu{}^{(+)}(t) + \mathbf{p}_\mu{}^{(-)}(t), \tag{109}$$

where ω_λ is the frequency in the unloaded cavity. It will become evident below, that in a laser we have $\omega \approx \nu$, and that the remaining amplitudes $E^{(\pm)}$, $\mathbf{p}^{(\pm)}$ vary much more slowly than $\exp(i\omega t)$. We therefore neglect $\dot{\tilde{E}}_\lambda$ compared to $\omega_\lambda \tilde{E}_\lambda$, and $\dot{\tilde{\mathbf{p}}}_\mu$ compared to $\nu \tilde{\mathbf{p}}_\mu$. Thus we obtain

$$\dot{\tilde{E}}_\lambda^{(-)} + \kappa_\lambda \tilde{E}_\lambda^{(-)} = -2\pi i \omega_0 \sum_\mu \mathbf{U}_\lambda(\mathbf{x}_\mu)\, \tilde{\mathbf{p}}_\mu^{(-)} \exp[i(\nu_\mu - \omega_\lambda)t\,], \tag{110}$$

where $\omega_0 \approx \omega_\lambda \approx \nu_\mu$. Using

$$E_\lambda^{(\mp)} = \exp(\pm i\omega_\lambda t)\, \tilde{E}_\lambda^{(\mp)};\ \mathbf{p}_\mu^{(\mp)} = \tilde{\mathbf{p}}_\mu^{(\mp)} \exp(\pm i\nu_\mu t), \tag{111}$$

we obtain, finally,

$$\dot{E}_\lambda^{(-)} = (i\omega_\lambda - \kappa_\lambda)\, E_\lambda^{(-)} - i\omega_0 2\pi \sum_\mu \mathbf{p}_\mu^{(-)}\, \mathbf{U}_\lambda(\mathbf{x}_\mu). \tag{112}$$

Similarly we find

$$\dot{\mathbf{p}}_\mu^{(-)} = (i\nu_\mu - \gamma)\, \mathbf{p}_\mu^{(-)} + i \frac{|\boldsymbol{\theta}_{12}|^2}{\hbar} \sum_\lambda \mathbf{U}_\lambda(\mathbf{x}_\mu)\, E_\lambda^{(-)}\, d_\mu, \tag{113}$$

where γ is the atomic line half width, and

$$\dot{d}_\mu = \frac{d_{0,\mu} - d_\mu}{T} + \frac{2i}{\hbar} \left\{ \left(\sum_\lambda E_\lambda^{(+)}\, \mathbf{U}_\lambda(\mathbf{x}_\mu) \right) \mathbf{p}_\mu^{(-)} - \left(\sum_\lambda E_\lambda^{(-)}\, \mathbf{U}_\lambda(\mathbf{x}_\mu)\, \mathbf{p}_\mu^{(+)} \right) \right\}. \tag{114}$$

where we have neglected terms $\sim \exp(\pm 2i\omega t)$ which are small compared to unity (rotating wave approximation). The eqns (112), (113) and (114) again represent a complete set of laser equations.

3.4. DIMENSIONLESS QUANTITIES

In these lectures we shall use a notation which makes contact directly with the quantum theoretical treatment by putting

$$E_\lambda^{(-)} = -i\sqrt{(2\pi\hbar\omega_\lambda)}\, b_\lambda^* \tag{115}$$

$$E_\lambda^{(+)} = i\sqrt{(2\pi\hbar\omega_\lambda)}\, b_\lambda. \tag{116}$$

In a fully quantum mechanical treatment, b_λ^*, b_λ are creation and annihilation operators. Here, they are merely time dependent, complex *dimensionless* amplitudes.

We decompose the atomic polarization $\mathbf{p}^{(+)}$ into a quantity with the dimension of a dipole moment (ex_{21}) and a remaining dimensionless factor ρ_{21}, thus

$$\mathbf{p}_\mu^{(+)} = \boldsymbol{\theta}_{21} \cdot \rho_{21,\mu}, \tag{117}$$

and correspondingly

$$\mathbf{p}_\mu^{(-)} = \boldsymbol{\theta}_{12} \cdot \rho_{12,\mu}. \tag{118}$$

Further, we write

$$N_{2,\mu} = \rho_{22,\mu}, \tag{119}$$

and

$$N_{1,\mu} = \rho_{11,\mu}. \tag{120}$$

In the quantum theoretical treatment, ρ_{21}, ρ_{12}, etc., are the elements of the density matrix of the two-level atom. Both in that treatment and this, they are time dependent classical functions. Finally, we introduce the coupling constant

$$g_{\mu\lambda} = i\boldsymbol{\theta}_{21}\sqrt{\left(\frac{2\pi\omega_\lambda}{\hbar}\right)}\,\mathbf{U}_\lambda(\mathbf{x}_\mu). \tag{121}$$

By use of (115), (116), (117), (118), (119) and (120), the original eqns (112), (113) and (114) acquire the form given in the next section which we shall use in the subsequent sections.

3.5. THE SEMICLASSICAL LASER-EQUATIONS[15]

The complete set of equations for two-level atoms is given by

$$\dot{b}_\lambda^*(t) = (i\omega_\lambda - \kappa_\lambda)\, b_\lambda^*(t) + i\sum_\mu g_{\mu,\lambda}^*\, \rho_{12,\mu}(t), \tag{122}$$

$$\dot{\rho}_{12,\mu}(t) = (iv_{21,\mu} - \gamma_{21})\, \rho_{12,\mu} - i\sum_\lambda g_{\mu,\lambda}\, b_\lambda^*(\rho_{22} - \rho_{11})_\mu, \tag{123}$$

$$\dot{d}_\mu = \frac{d_{0,\mu} - d_\mu}{T} + 2i\rho_{21,\mu}\sum_\lambda g_{\mu,\lambda}\, b_\lambda^* - 2i\rho_{12,\mu}\sum_\lambda g_{\mu,\lambda}^*\, b_\lambda, \tag{124}$$

where

$$d_{0,\mu} = (w_{12} - w_{21}) / (w_{12} + w_{21}) \tag{125}$$

is the unsaturated inversion of a single atom and

$$(1/T) = w_{12} + w_{21} \tag{126}$$

is the relaxation time of the inversion (compare (18)); the inversion of atom μ is $d_\mu = (\rho_{22} - \rho_{11})_\mu$; and κ_λ is the cavity half-width of mode λ.

If we retain the mode coupling caused by the conductivity σ—see (103) and (104)—we have to add the terms to the right-hand side of eqn (122)

$$\sum_{\lambda'} \sigma_{\lambda\lambda'}\sqrt{(\omega_{\lambda'}/\omega_\lambda)}\, b_{\lambda'}^*. \tag{122a}$$

These terms are of fundamental importance for the treatment of ultrashort pulses generated by loss-modulation. In this case, $\sigma_{\lambda\lambda'}$ is assumed to be non-vanishing at least for neighbouring axial modes, and to be modulated at a frequency close to the axial mode frequency spacing.

For the derivation of the above equations it was implicitly assumed that the upper laser level is pumped directly from the lower laser level. In reality, the pumping is achieved via auxiliary levels. We indicate how the equations must be altered for a pumping scheme given in Fig. 3. For the sake of completeness, we write down all equations. The laser action is assumed to occur between levels 2 and 1. The levels 0 and 3 serve as auxiliary levels for the pumping process.

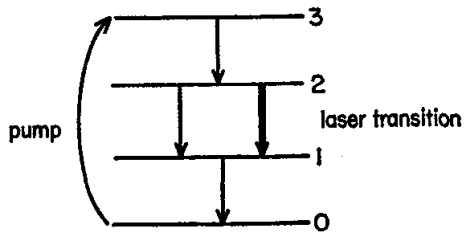


FIG. 3

$$\dot{b}_\lambda^* = (i\omega_\lambda - \kappa_\lambda)\, b_\lambda^* + i\sum_\mu g_{\mu,\lambda}^*\, \rho_{12,\mu} \tag{127}$$

$$\dot{\rho}_{12,\mu} = (iv_\mu - \gamma)\, \rho_{12,\mu} - i\sum_\lambda g_{\mu,\lambda}(N_{2,\mu} - N_{1,\mu})\, b_\lambda^* \tag{128}$$

$$\dot{N}_{0,\mu} = N_{1,\mu}\, w_{10} - N_{0,\mu}\, w_{03} \tag{129}$$

$$\dot{N}_{1,\mu} = N_{2,\mu}\, w_{21} - N_{1,\mu}\, w_{10} - \left\{ i\rho_{21,\mu}\sum_\lambda g_{\mu,\lambda}\, b_\lambda^* - i\rho_{12,\mu}\sum_\lambda g_{\mu,\lambda}^*\, b_\lambda \right\} \tag{130}$$

$$\dot{N}_{2,\mu} = N_{3,\mu}\, w_{32} - N_{2,\mu}\, w_{21} + \{\ldots\} \tag{131}$$

$$\dot{N}_{3,\mu} = N_{0,\mu}\, w_{03} - N_{3,\mu}\, w_{32}. \tag{132}$$

The $N_{i,\mu}$'s are the occupation numbers of the ith level of the μth atom and w_{ik} is the transition rate from level i to level k caused by incoherent sources (pump, spontaneous decay, radiationless transitions).

3.6. DERIVATION OF RATE EQUATIONS[16]

Before we discuss the solution of the semiclassical equation we shall demonstrate how to obtain the rate equations which we have used in Section 2. We put $b_\lambda^* = B_\lambda^* e^{(i\Omega_\lambda t)}$ where Ω_λ is the frequency of mode λ in the loaded cavity

and assume that B_λ^* changes only slowly with time (compared to γ) and also that $N_{2,\mu}-N_{1,\mu}$ changes slowly. The integration of (128) then yields

$$\rho_{12,\mu} = \sum_\lambda \frac{-ig_{\mu,\lambda}(N_{2,\mu}-N_{1,\mu})b_\lambda^*}{i(\Omega_\lambda-\nu_\mu)+\gamma}. \tag{133}$$

We insert (133) into eqn (127), multiply this equation by b_λ, and add the complex conjugate expression to the result. This gives us, finally

$$\dot{n}_\lambda = -2\kappa_\lambda n_\lambda + \sum_\mu \sum_{\lambda'} \left[\frac{g_{\mu,\lambda}^* g_{\mu,\lambda'}(N_{2,\mu}-N_{1,\mu}) b_{\lambda'}^* b_\lambda}{i(\Omega_{\lambda'}-\nu_\mu)+\gamma} + \text{c.c.} \right], \tag{134}$$

where $n_\lambda = b_\lambda^* b_\lambda$ is the photon number.

We shall neglect all terms with $b_{\lambda'}^* b_\lambda (\lambda \neq \lambda')$. This can be justified as follows. As we shall show in Section 4, the phases of the b^*'s fluctuate. As long as no phase locking occurs the phase fluctuations of the different b's are uncorrelated. Thus the mixed terms $(\lambda \neq \lambda')$ on the right hand side of eqn (134) vanish, if an average over the phases is taken. The *rate equations for the photon numbers* read thus

$$\dot{n}_\lambda = -2\kappa_\lambda n_\lambda + \sum_\mu \frac{2\gamma |g_{\mu,\lambda}|^2}{(\Omega_\lambda-\nu_\mu)^2+\gamma^2} (N_{2,\mu}-N_{1,\mu}) n_\lambda. \tag{135}$$

Equations (129) and (133) have already the required form of equations between occupation numbers.

When we insert (132) into (130) and (131) and apply the same reasoning, we find

$$\dot{N}_1 = N_{2,\mu} w_{21} - N_{1,\mu} w_{10} + \sum_\lambda \frac{2\gamma |g_{\mu,\lambda}|^2}{(\Omega_\lambda-\nu_\mu)^2+\gamma^2} (N_{2,\mu}-N_{1,\mu}) n_\lambda \tag{136}$$

$$\dot{N}_2 = N_{3,\mu} w_{32} - N_{2,\mu} w_{21} - \sum_\lambda \frac{2\gamma |g_{\mu,\lambda}|^2}{(\Omega_\lambda-\nu_\mu)^2+\gamma^2} (N_{2,\mu}-N_{1,\mu}) n_\lambda. \tag{137}$$

The total set of rate equations is represented by eqns (135), (129), (132), (136) *and* (137). The rate equations allow us to determine photon numbers and atomic occupation numbers; they do not contain any information about frequency shifts. For this reason the frequencies in the unloaded cavity, ω_λ are usually used.

3.7. METHOD OF SOLUTION FOR THE STATIONARY STATE[17]

We now come back to the general equations (127)–(132). Because exact solutions of them are known only for special cases, e.g. single mode operation, we present an iteration which exhibits some of the important new features of

the semiclassical equations. We treat explicitly the two-level system described by eqns (122), (123) and (124), although the procedure is quite general. The spirit of this approach is the following. In the field equations (122) the polarization P acts as a source term, which in turn is determined as a function of the field, $b_\lambda(t)$, via the matter equations. We determine this dependence of P on b_λ approximately and insert the resulting expression into the field equation (122), so that we obtain an equation for the field amplitude alone.

We assume that by the pumping, as well as by relaxation processes, an inversion given by

$$d_\mu^{(0)} = d_{\mu,0} \tag{138}$$

is established without laser action in zero'th approximation. We assume further that due to the laser process, coherent fields of the form

$$b_\lambda^* = B_\lambda^* \exp(i\Omega_\lambda t) \tag{139}$$

are built up. The ansatz (139) would suggest itself, if the laser field were a classical field. Since it is generated by quantum processes, the ansatz (139) requires a deeper motivation, which will be given in the fully quantum mechanical treatment in Section 4.

The complex amplitude B_λ^* and the frequency Ω_λ are unknown parameters which are to be determined in a self-consistent way. The frequency Ω_λ is assumed to be real, because we confine ourselves in the present paragraph to the stationary state. Under certain circumstances B_λ^* may not be completely time independent but may vary very slowly.

Before we treat the general case of the multimode equations, (122)–(124), we give an example for a single mode. We assume resonance, i.e. $\omega = \nu$ (in which case one may assume in a self-consistent manner $\Omega = \omega$), and assume further, that g, the coupling constant is independent of μ and λ, and g is real. Inserting (138) and (139) into (123) we find the atomic polarization in first order

$$\rho_{12,\mu}^{(1)}(t) = -id_0 B^* \exp(i\omega t)\, g/\gamma.$$

Inserting this result into eqn (124), we may integrate this equation in order to find an improved value for the inversion given by

$$d_\mu = d_0 - 4d_0|B|^2(g^2T/\gamma).$$

We now determine an improved dipole moment, by inserting the new expression for d_μ into eqn (123). The integration of this equation yields immediately

$$\rho_{12,\mu}^{(2)}(t) = -id_0 B^* \exp(i\omega t)\frac{g}{\gamma}(1 - 4|B|^2 g^2 T/\gamma).$$

Thus the dipole moment is known as a function of the field. The field in turn is determined by the polarization (or dipole moments). Thus we introduce $\rho_{12}^{(2)}$ into the field equation (122) and obtain the following self-consistent field equation

$$\dot{b}^* = (i\omega - \kappa)\, b^* + d_0\, N \frac{g^2}{\gamma} b^* (1 - 4|b|^2 g^2 T/\gamma),$$

which is a nonlinear equation for b^*. The total number of atoms is N. The above steps can be represented by the following scheme.

field $b^* = B^* \exp(i\omega t)$ and inversion $d_\mu = d_0$ given $\Big\}$ (123) $\Rightarrow$ dipole moment, $\rho_{12} \sim b^*$ $\Big\}$ (124) $\Rightarrow$ $\Downarrow$

$\Uparrow$ inversion $\underbrace{d_\mu = d_0 - \ldots |b|^2}_{(123)}$

new field equation $\Leftarrow$ (122) $\Big\{$ improved dipole moment $\rho_{12} \sim b^* + \ldots |b|^2 b^*$ $\Leftarrow$ $\Downarrow$

The effective field equation is nonlinear in the amplitude. For the whole laser theory this nonlinearity is of utmost importance. To see this, we insert $b^* = B^* \exp(i\omega t)$ into this field equation and obtain

$$\kappa = d_0\, N \frac{g^2}{\gamma} (1 - 4|b|^2 g^2 T/\gamma),$$

from which we may determine the photon number $n = |b|^2$ as a function of the unsaturated inversion d_0. If we had neglected the nonlinear term, no steady state would arise at all. We now treat the general case of eqns (122) and (123), using again the ansatz (138) and (139).

The atomic dipole moments are changed under the influence of a given light field according to eqn (123) whose integration yields[18]

$$\rho_{12,\mu}(t) = -d_{\mu,0} \sum_\lambda B_\lambda^*\, g_{\mu\lambda} \frac{\exp(i\Omega_\lambda t)}{(\Omega_\lambda - \nu_\mu - i\gamma)}. \tag{140}$$

Inserting this result into (124) and integrating this equation we obtain as an

improved value for the inversion

$$d_\mu = d_{\mu,0} + \left\{ 2id_{0,\mu} \sum_{\lambda\lambda'} B_\lambda B^*_{\lambda'} g^*_{\mu\lambda} g_{\mu\lambda'} \frac{\exp[i(\Omega_{\lambda'} - \Omega_\lambda)t]}{(\Omega_{\lambda'} - \nu_\mu - i\gamma)\{1/T + i(\Omega_{\lambda'} - \Omega_\lambda)\}} - \text{c.c.} \right\} \tag{141}$$

Using the improved inversion we can obtain further improvement in the atomic dipole moment. For this purpose we insert (141) into (123) and obtain after the integration

$$\rho^{(2)}_{12,\mu} = -d_{\mu,0} \sum_\lambda B_\lambda^* g_{\mu\lambda} \frac{\exp(i\Omega_\lambda t)}{(\Omega_\lambda - \nu_\mu - i\gamma)} + 2d_{\mu,0} \sum_{\lambda\lambda'\lambda''} B_\lambda B^*_{\lambda'} B^*_{\lambda''} g^*_{\mu\lambda} g_{\mu\lambda'} g_{\mu\lambda''}$$
$$\times \frac{\exp[i(\Omega_{\lambda'} + \Omega_{\lambda''} - \Omega_\lambda)t]}{i\{(1/T) + i(\Omega_{\lambda'} - \Omega_\lambda)\}(\Omega_{\lambda'} - \Omega_\lambda + \Omega_{\lambda''} - \nu_\mu - i\gamma)}$$
$$\times \left(\frac{1}{\Omega_{\lambda'} - \nu_\mu - i\gamma} - \frac{1}{\Omega_\lambda - \nu_\mu + i\gamma} \right). \tag{142}$$

Equation (142) expresses the atomic dipole moments and thus the polarization as an explicit function of the field amplitude b_λ.

We now take into account the reaction of the dipole moments on the light field. Therefore we insert the $\rho^{(2)}_{12,\mu}$'s, which have been obtained above, in the field equations instead of $\rho_{12,\mu}$. This yields

$$b_\lambda^* = (i\omega_\lambda - \kappa) b_\lambda^* - i \sum_\mu d_{0,\mu} |g_{\mu\lambda}|^2 \frac{B_\lambda^* \exp(i\Omega_\lambda t)}{(\Omega_\lambda - \nu_\mu - i\gamma)}$$
$$+ 2 \sum_{\mu\lambda_1\lambda_2\lambda_3} d_{0,\mu} B_{\lambda_1} B^*_{\lambda_2} B^*_{\lambda_3} g^*_{\mu\lambda} g^*_{\mu\lambda_1} g_{\mu\lambda_2} g_{\mu\lambda_3}$$
$$\times \frac{\exp[i(\Omega_{\lambda_2} + \Omega_{\lambda_3} - \Omega_{\lambda_1})t]}{[(1/T) + i(\Omega_{\lambda_2} - \Omega_{\lambda_1})][\Omega_{\lambda_2} + \Omega_{\lambda_3} - \Omega_{\lambda_1} - \nu_\mu - i\gamma]}$$
$$\times \left(\frac{1}{\Omega_{\lambda_2} - \nu_\mu - i\gamma} - \frac{1}{\Omega_{\lambda_1} - \nu_\mu + i\gamma} \right). \tag{143}$$

It can be shown that the above iteration procedure is justified if the atomic linewidth γ is larger than the other frequencies inherent in the problem.[18] Equation (143) represents the basis of the following two chapters. In fact, the whole further analysis consists in performing the summations and integrations over energies and atomic indices, and in solving eqn (143). This equation describes the oscillation of the modes taking into account their interaction through the response of the atomic system.

According to different combinations $\Omega_{\lambda_2}+\Omega_{\lambda_3}-\Omega_{\lambda_1}$ on the right hand side of (143), new frequencies may occur. Thus, by the cooperation of two or three fields an additional field mode may be created. This kind of frequency mixing is related to those in inactive materials. To exhibit some further features of eqns (143) we specialize them for single and two-mode operation.

a. Single mode operation

In this case, we have

$$\kappa+i(\Omega-\omega) = -i\sum_{\mu}\frac{|g_\mu|^2 d_{0,\mu}}{(\Omega-\nu_\mu-i\gamma)}+\sum_{\mu}\frac{B^*B|g_\mu|^4 d_{0,\mu}4i\gamma\mathrm{T}}{[\Omega-\nu_\mu-i\gamma][(\Omega-\nu_\mu)^2+\gamma^2]}. \tag{144}$$

We split eqn (144) into its real and imaginary parts.
The *real part* is

$$2\kappa = G_u - Sn, \tag{145}$$

where

$$G_u = \sum_{\mu}\frac{d_{0\mu}|g_\mu|^2 2\gamma}{(\Omega-\nu_\mu)^2+\gamma^2}, \tag{146}$$

and

$$S = \sum_{\mu}\frac{d_{0\mu}|g_\mu|^4 8\gamma^2 T}{[(\Omega-\nu_\mu)^2+\gamma^2]^2}. \tag{147}$$

After multiplication with the number of photons n on both sides of (145), the left-hand side represents the loss rate of photons, whereas the first sum on the right-hand side gives the increase of photon number due to induced emission from atoms with a given inversion.

If γ is small compared to the spread of atomic energies, $(\gamma/\{(\Omega-\nu_\mu)^2+\gamma^2\})$ can be approximated by the δ-function, $\delta(\Omega-\nu_\mu)$, representing energy conservation. Only those atoms whose transition energy coincides with the mode frequency within a width γ participate in the process of stimulated emission. The second sum Sn takes into account that the inversion is decreased by the laser process. This effect is called "hole burning into the gain curve".
The *imaginary part* of (144) is

$$(\Omega-\omega) = (\Delta\Omega)_u + n(\Delta\Omega)_s, \tag{148}$$

where

$$(\Delta\Omega)_u = -\sum_{\mu}\frac{|g_\mu|^2(\Omega-\nu_\mu)\,d_{0\mu}}{(\Omega-\nu_\mu)^2+\gamma^2} \tag{149}$$

and

$$(\Delta\Omega)_s = 4\gamma T\sum_{\mu}\frac{|g_\mu|^4 d_{0\mu}(\Omega-\nu_\mu)}{[(\Omega-\nu_\mu)^2+\gamma^2]^2} \tag{150}$$

and the subscripts u and s denote the unsaturated and saturated conditions respectively.

According to the imaginary part, the active laser atoms change the frequency ω of the light mode in the unloaded cavity. The first sum on the right hand side has the same form as the usual dispersion formula with a given occupation of the atomic system. This expression could have also been obtained by second order perturbation theory. The only difference is that we have already used a renormalized frequency Ω instead of ω as occurring in the usual perturbation procedure. What is essentially new, however, is represented by the occurrence of the second sum. In it, the square of the light amplitude, or correspondingly, the number of photons occurs. This term therefore describes the change of the dispersion due to the adjustment of the atomic occupation to the intensity of the light field. It should be noted that this response of the atomic system is time independent. In the steady state case, the photon number adjusts in such a way that gain and loss compensate each other as described by eqn (145). With the photon number fixed in this way we can calculate the dispersion part of the right hand side of eqn (148), which leads immediately to an expression for the frequency shift in the inverted system as compared to the unloaded cavity.

b. Two mode operation, general features

Specialization of (143) yields

$$\kappa+i(\Omega_\lambda-\omega_\lambda)=\sum_\mu \frac{d_{0\mu}|g_{\mu\lambda}|^2}{\gamma+i(\Omega_\lambda-\nu_\mu)} -4\gamma T\sum_{\mu,\lambda'}\frac{d_{0\mu}\,B_{\lambda'}{}^*\,B_{\lambda'}|g_{\mu\lambda}|^2|g_{\mu\lambda'}|^2}{[\gamma+i(\Omega_\lambda-\nu_\mu)][\gamma^2+(\Omega_{\lambda'}-\nu_\mu)^2]}$$

$$+2\sum_{\substack{\mu\\(\lambda'\neq\lambda)}}\frac{d_{0\mu}\,B_{\lambda'}{}^*\,B_{\lambda'}|g_{\mu\lambda}|^2|g_{\mu\lambda'}|^2}{[(1/T)+i(\Omega_\lambda-\Omega_{\lambda'})][\Omega_\lambda-\nu_\mu-i\gamma]}$$

$$\times\left(\frac{1}{\Omega_\lambda-\nu_\mu-i\gamma}-\frac{1}{\Omega_{\lambda'}-\nu_\mu+i\gamma}\right) \tag{151}$$

for $\lambda = 1$ and 2. Whereas the left-hand side and the first sum on the right-hand side are the same as for single mode operation, new features occur due to the second and third sum. These arise from a time-independent and time dependent atomic response.

The second sum gives rise to a change both of gain and dispersion of the first mode caused by the photon numbers of the first and the second mode. Whereas the change of the gain is proportional to

$$\frac{n_{\lambda'}\gamma}{[\gamma^2+(\Omega_{\lambda'}-\nu_\mu)^2]}, \tag{152}$$

the change of the dispersion is proportional to

$$\frac{n_{\lambda'}(\Omega_{\lambda'}-\nu_\mu)}{[\gamma^2+(\Omega_{\lambda'}-\nu_\mu)^2]}. \tag{153}$$

If we take also into account the fact that the main contributions of the sums over μ stem from $\nu_\mu \approx \Omega_{\lambda'}$ we see that the mutual influence of both modes has a longer range for the dispersion than for the gain. In fact, if $|\Omega_{\lambda'}-\Omega_\lambda|$ is bigger than γ there is practically no influence of one photon number, n_λ, on the other, $n_{\lambda'}$. On the other hand, the "hole burning" changes the dispersion. Whereas the hole burning stems from the time independent atomic response, the time dependent atomic response gives rise to the third sum in (151). This can be seen by following up the iteration steps performed above. Under certain circumstances it leads also to an appreciable frequency pushing of the two modes.

In the following we give only few examples how eqn (143) may be solved. For a systematic treatment see the article of the present author in Encyclopedia of Physics.[1]

3.8. THE SOLID STATE LASER WITH A HOMOGENEOUSLY BROADENED LINE

Single and multimode laser action[19]

The eqns (122), (123) and (124) or the eqn (143) allow us to treat two problems: (1) the determination of the mean number of photons in the laser modes; (2) the determination of the actual laser frequencies. Although in general the solid state laser will have an inhomogeneously broadened line, we treat first the simpler case of a homogeneously broadened line which allows us, incidentally, to elucidate the general procedure for the solution of eqn (143).

a. Single mode operation

We start with the eqn (144) where we have to perform the sums (or integrals) over μ. Because in the present case the atomic transition frequency ν does not depend on the atomic index μ, no integration over such frequencies is required. On the other hand we still have to sum up over the random atomic positions $\mathbf{x}_\mu$. This sum can be performed to a very good approximation by an integration over $\mathbf{x}$,

$$\sum_\mu \ldots = \rho \int \ldots \mathrm{d}^3 V, \tag{154}$$

where ρ is the density of the laser active atoms. In the following we assume that the unsaturated inversion, $d_{0\mu} = d_0$ does not depend on the position $\mathbf{x}_\mu$, i.e. we assume a spatially homogeneous pumping (which is also time independ-

ent, of course). In order to perform the integration (154), we adopt, for the coupling constant $g_{\mu,\lambda}$ between the light mode λ and the atom μ the form

$$g_{\mu\lambda} = i\left(\frac{4\pi\omega_\lambda}{\hbar V}\right)^{\frac{1}{2}} \mathbf{e}_\lambda \boldsymbol{\theta}_{21} \sin(k_\lambda x_\mu), \tag{155}$$

corresponding to purely axial modes where $\boldsymbol{\theta}_{12}$ is the atomic dipole moment, and V is the volume of the active material. The integration for modes with an arbitrary **k**-direction can be done in an analogous way.

We multiply (144) on both sides with $\gamma + i(\Omega - \nu)$. The real part of the new equation then reads:

$$\gamma\kappa - (\Omega-\omega)(\Omega-\nu) = \frac{2\pi\nu^2|\theta_{12}|^2\rho d_0}{\hbar\omega}\left[1 - \frac{12\pi\nu^2|\theta_{12}|^2\gamma T\bar{n}}{\hbar\omega\{\gamma^2+(\Omega-\nu)^2\}}\right] \tag{156}$$

and the imaginary part is given by

$$\Omega = \frac{\gamma\omega + \kappa\nu}{\gamma+\kappa}. \tag{157}$$

Equation (156) is identical with a rate equation, which determines the photon density $\bar{n}$ as a function of the pumping (via d_0). The photon density, $\bar{n}$, is defined as $B^* B/V$. Solving (156) for $\bar{n}$ yields

$$\bar{n} = \frac{d_{\text{thr}}(\Delta)\,\rho}{6T\kappa}\left(1 - \frac{d_{\text{thr}}(\Delta)}{d_0}\right), \tag{158}$$

where

$$d_{\text{thr}}(\Delta) = \frac{2\hbar\gamma\kappa\omega}{4\pi\rho\nu^2|\theta_{12}|^2}\left(1 + \frac{\Delta^2}{(\gamma+\kappa)^2}\right) \tag{159}$$

and $\Delta = \omega - \nu$. From (159) the laser condition (13) follows.

Laser action starts if $\bar{n}$ becomes positive, or according to (159), if the atomic inversion d_0, which is fixed by the pumping and the incoherent decay processes, becomes bigger than d_{thr}, that is, if we have

$$d_0 \geqslant d_{\text{thr}}, \tag{160}$$

or, explicitly[20]

$$\frac{D_0}{V} \equiv \frac{N(N_2-N_1)_0}{V} \geqslant \frac{2\hbar\gamma\kappa\omega}{4\pi\nu^2|\theta_{12}|^2}\left(1 + \frac{\Delta^2}{(\gamma+\kappa)^2}\right). \tag{161}$$

In this equation, N is the total number of atoms; $(N_2-N_1)_0$ is the atomic inversion of a single atom (without laser action); and D_0 is the total (unsaturated) inversion.

The laser frequency Ω given by eqn (157) is shifted from the frequency Ω in the unloaded cavity to the centre of the line[21] (frequency pulling). For a homogeneously broadened line, this shift does not depend on pump power.

b. Multiple mode operation

The general solution of the eqn (143) for the multiple mode case is difficult and has not been found yet, (at least to our knowledge†). Therefore, we discuss only the most important feature of this case. As has been explained in 3.7, the light modes cause both a time dependent and a time independent response of the atomic system. Provided the spacing between the frequencies of the different modes is big enough,

$$|\Omega_{\lambda_1}-\Omega_{\lambda_2}|>1/T, \qquad \lambda_1\neq\lambda_2, \tag{162}$$

the time dependent response can be neglected. We neglect further, the possibility of frequency mixing, an effect which is, anyhow, very small. With these simplifications the real and imaginary parts of eqn (143) lead to two sets of equations, which determine the photon density of the modes and the frequency shift.

α. Equations for the photon densities of M modes from the real part

$$\begin{aligned}
&\tfrac{3}{2}\bar{n}_1\frac{\kappa_1}{d_{\text{thr}}(\Delta_1)}+\bar{n}_2\frac{\kappa_2}{d_{\text{thr}}(\Delta_2)}+\ldots+\bar{n}_m\frac{\kappa_m}{d_{\text{thr}}(\Delta_m)}=\frac{\rho}{4T}\left(1-\frac{d_{\text{thr}}(\Delta_1)}{d_0}\right)\\
&\bar{n}_1\frac{\kappa_1}{d_{\text{thr}}(\Delta_1)}+\tfrac{3}{2}\bar{n}_2\frac{\kappa_2}{d_{\text{thr}}(\Delta_2)}+\ldots+\bar{n}_m\frac{\kappa_m}{d_{\text{thr}}(\Delta_m)}=\frac{\rho}{4T}\left(1-\frac{d_{\text{thr}}(\Delta_2)}{d_0}\right)\\
&(m\text{ equations}) \qquad\qquad \vdots
\end{aligned} \tag{163}$$

where ω is to be supplemented with the index λ and $d_{\text{thr}}(\Delta_\lambda)$ is defined in (159). These equations are identical with the rate equations (see Section 2) and have been discussed in 2.7.

β. Equations for the frequency shift

From the imaginary part of eqn (143), using (163) we get

$$\Omega_\lambda=\frac{\gamma\omega_\lambda+\kappa_\lambda\nu}{\gamma+\kappa_\lambda}, \tag{164}$$

i.e. all modes show the power independent pulling, known from single mode action.

† A case of particular interest, which has been treated, is that of mode locking in a multimode laser.

3.9. THE SOLID STATE LASER WITH AN INHOMOGENEOUSLY BROADENED GAUSSIAN LINE[22]

The essential difference between the preceding Section 3.8 and the present one is that we must not only sum up over the atomic positions x_μ but also over the transition energy (or transition frequency ν_μ).

a. One mode

As an example we choose a Gaussian distribution for ν. It should be noted that other distributions can be treated similarly, or in the case of a Lorentzian line shape, even more simply. The sums over the atomic indices then lead to the following expressions

$$\kappa+i(\Omega-\omega)=\frac{\rho A d_0}{2}\int\left[\frac{\gamma-i(\Omega-\nu)}{\gamma^2+(\Omega-\nu)^2}-3\gamma T A\bar{n}\frac{\gamma-i(\Omega-\nu)}{\{\gamma^2+(\Omega-\nu)^2\}^2}\right]\times\frac{\exp[-\{(\nu-\nu_0)/\alpha\}^2]\mathrm{d}\nu}{\alpha\sqrt{\pi}}, \tag{165}$$

where

$$A=4\pi\nu_0^2|\theta_{12}|^2/\hbar\omega,$$

α is the inhomogeneous atomic half-width, d_0 is the unsaturated atomic inversion, and ν_0 is the frequency at the line centre. In eqn (165) we have already integrated over $\mathbf{x}$, and it was assumed that the vector of polarization $\mathbf{e}$ is parallel to the atomic dipole moment $\boldsymbol{\theta}_{12}$. The integral over ν is evaluated under the assumption that the inhomogeneous line width is large compared to the homogeneous Lorentzian line width, which is described by the imaginary part, γ, of ν. Using the result of the evaluation of the integral and splitting eqn (165) into real and imaginary parts, we find the following equations for the frequency shift and for the photon density.

Equation for the frequency shift

$$\Omega-\omega=-\frac{\rho A d_0}{\alpha}\exp(-\delta^2)\,\Phi(\delta)+\bar{n}C, \tag{166}$$

where

$$\Phi(\delta)=\int_0^\delta \exp(+u^2)\,\mathrm{d}u \tag{167}$$

$$C=\{3A^2\rho d_0 T\,\pi^{\frac{1}{2}}\,\delta\exp(-\delta^2)\}/4\alpha^2 \tag{168}$$

and

$$\delta=(\Omega-\nu_0)/\alpha. \tag{169}$$

The total number of atoms is N, and α is the Doppler width (compare eqn (165)). The first term on the right hand side of (166) is peculiar to an inhomogeneous Gaussian line broadening.[23] Note that this term is pump dependent on account of its factor d_0. It gives rise to a *pulling* of the *mode frequency* ω in the unloaded cavity to the centre of the emission line, ν_0. The next term depends on the *photon density* (or, in other words, on the *field intensity* in the cavity) and describes a frequency pushing. In order to determine its size, we insert the value of $\bar{n}$, which is anticipated from eqn (173) below and we obtain

$$\Omega-\omega = -\kappa\frac{2d_0}{d_{\text{thr}}(\delta)\,\pi^{\frac{1}{2}}}\left[\Phi(\delta)-\delta\frac{\gamma}{\alpha}\pi^{\frac{1}{2}}\left(1-\frac{d_{\text{thr}}(\delta)}{d_0}\right)\right], \tag{170}$$

where $d_{\text{thr}}(\delta)$ denotes the threshold inversion, which is given by

$$d_{\text{thr}}(\delta) = \frac{\hbar\alpha\kappa\omega\exp\delta^2}{2(\pi^3)^{\frac{1}{2}}\rho|v_0|^2|\theta_{12}|^2}. \tag{171}$$

From (170) we see that the repulsive term is small compared to the first on account of its factor γ/α.

Equation for the photon density.

This follows from the real part of (165) and reads

$$\kappa = \frac{\rho A d_0}{2}\left[\frac{\pi^{\frac{1}{2}}}{\alpha}\exp(-\delta^2)-3AT\bar{n}\frac{\pi^{\frac{1}{2}}\exp-\delta^2}{2\gamma\alpha}\right] \tag{172}$$

Solving this equation for $\bar{n}$ we arrive at

$$\bar{n} = \frac{2\gamma}{3AT}\left(1-\frac{d_{\text{thr}}(\delta)}{d_0}\right) = \frac{\gamma}{\alpha}\frac{\pi^{\frac{1}{2}}\rho d_{\text{thr}}(0)}{3\kappa T}\left(1-\frac{d_{\text{thr}}(\delta)}{d_0}\right). \tag{173}$$

This equation allows us to determine the photon-density (or light intensity) as a function of the pump rate, which occurs in d_0 and T (compare eqn (125) and (126)). Equation (172) can also be derived in the rate equation approximation (compare 2.7).

b. Two modes

A detailed discussion shows that the photon numbers of the two modes influence one another significantly only if the frequency separation $\Delta\Omega = \Omega_1-\Omega_2$ is of the order of the natural linewidth, γ. For $(\gamma/\Delta\Omega)^2<1$, we can therefore use the expression (173) for $\bar{n}_\lambda$. The coupling between the two photon numbers decreases roughly as $(\gamma/\Delta\Omega)^2$. On the other hand the mutual

influence of the frequencies is larger and decreases only as $(\gamma/\Delta\Omega)$ if $|\Delta\Omega| > \gamma$ in the same order of approximation.

Equations for the frequency shifts

$$\Omega_1 - \omega_1 = \frac{-\rho A d_0}{\alpha} \exp(-\delta_1{}^2)\, \Phi(\delta_1) + \bar{n}_1 C_1 + \bar{n}_2 M_{12}. \tag{174}$$

The first two sum terms are identical with those of the expression (166) for the frequency shift in single mode operation (we have merely to replace in (166) and in (169) δ by δ_1 and to put $C_1 = C$). The last sum term shows that the frequency shift of the first mode depends also on the intensity of the second mode. We quote the result for a large frequency separation $|\Delta\Omega| \gg \gamma$. In order to put the relative size of the various contributions in evidence, we rewrite (174) in the form (with $(\Delta\Omega)^2 \gg \gamma^2$),

$$\Omega_1 - \omega_1 = \kappa \frac{d_0}{d_{\text{thr}}(\delta_1)} \{g_1 + \bar{n}_1 g_1' + \bar{n}_2 g_2\} \tag{175}$$

where

$$g_1 = -\frac{2}{\pi^{\frac{1}{2}}} \Phi(\delta_1), \qquad g_1' = \frac{3AT\delta_1}{\alpha} \tag{176}$$

$$g_2 = \frac{2AT}{\Delta\Omega} \exp(\delta_1{}^2 - \delta_2{}^2). \tag{177}$$

The first two terms (176) are exactly the same as those of one mode, which, as we have discussed above, give rise to a pulling of the line toward the centre of the emission line. The following term (177) is brought about by a time independent depletion of the excited atomic states due to their interaction with the other mode. It is thus a hole burning effect as has been described in a macroscopic language by Bennett.[23] There is actually a further term in addition to g_2 which stems from the time dependent response of the atomic system. It is, however, in general small compared to g_2 in an inhomogeneously broadened line.

3.10. EQUATIONS OF MOTION FOR THE GAS LASER[24]

a. Haken–Sauermann treatment

The essential difference compared to the solid state laser is given here by the motion of the gas atoms. Consequently, the co-ordinate of a single atom is now given by $\mathbf{x}_\mu + \mathbf{v}_\mu t$. We allow further for arbitrary angles, $\theta_{\mu\lambda}$, between the vector of polarization of the light mode λ and the dipole moment of the μth

atom. The interaction constant $g_{\mu\lambda}$ between the mode λ and the atom μ takes thus the form

$$g_{\mu\lambda} = -\frac{ig}{2}\{\exp[i\,\mathbf{k}_\lambda\,(\mathbf{x}_\mu+\mathbf{v}_\mu\,t)]-\text{c.c.}\}\cos\theta_{\mu\lambda}. \tag{178}$$

The summation over μ runs in the corresponding equations over the positions $\mathbf{x}_\mu$, the velocities $\mathbf{v}_\mu$ (for which a Boltzmann distribution is assumed) and over all angles θ_μ. The equations of motion can now be taken directly from those referring to fixed atoms, (127)–(132) or (122)–(124), if the replacement (178) is made. The frequency ν_μ is simply to be identified with the centre-frequency ν_0 (the Doppler broadening is automatically taken care of by the explicit consideration of the atomic motion).

b. Lamb treatment

We represent still another way of introducing the atomic motion due to Lamb, which is different in form from, but completely equivalent to the procedure (178) of Haken and Sauermann outlined above. Consider an atom excited by some process at time t_0 to its upper level. Let the atom be at the position $\mathbf{x}_0$ at t_0 and have the velocity $\mathbf{v}$ so that at time $t > t_0$ the atom will be at

$$\mathbf{x} = \mathbf{x}_0+\mathbf{v}\,(t-t_0). \tag{179}$$

The corresponding density matrix $\rho(\mathbf{x}_0, t_0, \mathbf{v}; t)$ obeys the equation

$$\dot{\rho} = -\frac{i}{\hbar}[H, \rho]-\tfrac{1}{2}(\Gamma\,.\,\rho+\rho\,.\,\Gamma), \tag{180}$$

where

$$\Gamma = \begin{pmatrix}\gamma_a & 0\\ 0 & \gamma_b\end{pmatrix},$$

and H is the Hamiltonian of a single atom, γ_a, γ_b are the decay constants of the corresponding levels. The field in (180) has the argument as defined in (179), $H = H(\mathbf{x}_0+\mathbf{v}(t-t_0), t$. Furthermore, $\rho(\mathbf{x}_0, t_0, \mathbf{v}; t)$ has to obey the initial condition

$$\rho(\mathbf{x}_0, t_0, \mathbf{v}; t_0) = \begin{pmatrix}1 & 0\\ 0 & 0\end{pmatrix}. \tag{181}$$

Because the light mode interacts with a macroscopic polarization $\mathbf{P}(\mathbf{x}, t)$, we combine the contribution of all atoms which arrive at $\mathbf{x}$ at time t, no matter when or where they were excited. We note that

$$\mathbf{P}(\mathbf{x}, t) = (\boldsymbol{\theta}_{12}\,\bar{\rho}_{12}(\mathbf{x}, t)+\boldsymbol{\theta}_{21}\,\bar{\rho}_{21}(\mathbf{x}, t)), \tag{182}$$

where θ_{ij} is the dipole moment matrix element of a single atom, and $\bar{\rho}_{ij}$ is the density matrix element for the corresponding transition (i.e. the off-diagonal elements of the density matrix of the ensemble of atoms). Instead of deriving an expression for $\mathbf{P}(\mathbf{x}, t)$ we therefore derive $\bar{\rho}(\mathbf{x}, t)$. We assume that the excitation processes obey a Poisson distribution. Then $\bar{\rho}(\mathbf{x}, t)$ is obtained by integration over the time $(t - t_0)$ since the last excitation collision using the probability distribution $(1/T_p) \exp[-(t - t_0)/T_p]$, where T_p is the relaxation time for atomic inversion. We then find

$$\bar{\rho}(\mathbf{x}, t) = \frac{1}{T_p} \int_{-\infty}^{t} \exp[-(t-t_0)/T_p] \, \mathrm{d}t_0 \int \mathrm{d}V_0 \int \mathrm{d}v \tilde{\lambda}(\mathbf{x}_0, t_0, \mathbf{v}) \times \rho(\mathbf{x}_0, t_0, \mathbf{v}, t) \delta(\mathbf{x} - \mathbf{x}_0 - \mathbf{v}(t - t_0)), \tag{183}$$

where V_0, v are integration variables (space or velocity), and $\tilde{\lambda}(\mathbf{x}_0, t_0, \mathbf{v})$ is a mean excitation rate, whose significance becomes especially clear in eqn (188). Since $\rho(\mathbf{x}_0, t_0, \mathbf{v}, t)$ decays exponentially because of the Γ in eqn (180), the exponential function, $\exp[-(t-t_0)/T_p]$ can be neglected if $(1/T_p) < \gamma_a, \gamma_b$.

c. Equivalence between the formulation of Haken–Sauermann and Lamb

In order to demonstrate the equivalence of the formulation (180)–(183) with the formulation by means of eqns (127)–(132) where the matrix element (178) is used, we write $\bar{\rho}(\mathbf{x}, t)$ in the form

$$\bar{\rho}(\mathbf{x}, t) = \int \mathrm{d}V \bar{\rho}(\mathbf{x}, \mathbf{v}, t), \tag{184}$$

with

$$\bar{\rho}(\mathbf{x},\mathbf{v},t) = \frac{1}{T_p} \int_{-\infty}^{t} \exp[-(t-t_0)/T_p] \, \mathrm{d}t_0 \times \int \mathrm{d}V_0 \tilde{\lambda}(\mathbf{x}_0, t_0, \mathbf{v}) \, \rho(\mathbf{x}_0, t_0 \, \mathbf{v}, t) \delta(\mathbf{x} - \mathbf{x}_0 - \mathbf{v}(t - t_0)) \tag{185}$$

and investigate the time variation of $\bar{\rho}(\mathbf{x}, \mathbf{v}, t)$:

$$\frac{\mathrm{d}\bar{\rho}(\mathbf{x}, \mathbf{v}, t)}{\mathrm{d}t} = \frac{\hat{\lambda}}{T_p}(\mathbf{x}, t, \mathbf{v}) - \frac{1}{T_p} \bar{\rho} - \mathbf{v} \operatorname{grad}_{\mathbf{x}} \bar{\rho}(\mathbf{x}, \mathbf{v}, t) + \int_{-\infty}^{t} \mathrm{d}t_0 \exp[-(t-t_0)/T_p] \frac{\tilde{\lambda}}{T_p}(\mathbf{x} - \mathbf{v}(t-t_0), t_0, \mathbf{v}) \times \{-i/\hbar[H(\mathbf{x}, t), \rho(\mathbf{x} - \mathbf{v}(t-t_0), t_0, v, t)] - \tfrac{1}{2}(\Gamma\rho + \rho\Gamma)\} \tag{186}$$

where $\hat{\lambda} = \tilde{\lambda} \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix}$.

Evidently, H no longer contains t_0 so that it can be put in front of the integral.

Thus we find, finally

$$\frac{\mathrm{d}\bar{\rho}(\mathbf{x}, \mathbf{v}, t)}{\mathrm{d}t} + \mathbf{v}\,\mathrm{grad}_{\mathbf{x}}\,\bar{\rho}(\mathbf{x}, \mathbf{v}, t)$$

$$= \frac{\hat{\lambda}(\mathbf{x}, t, \mathbf{v}) - \bar{\rho}(\mathbf{x}, \mathbf{v}, t)}{T_p} - \frac{i}{\hbar}[H(\mathbf{x},t), \bar{\rho}(\mathbf{x}, \mathbf{v}, t)]$$

$$-\tfrac{1}{2}(\Gamma\bar{\rho}(\mathbf{x}, \mathbf{v}, t) + \bar{\rho}(\mathbf{x}, \mathbf{v}, t)\Gamma). \tag{187}$$

In the theory of fluids, (187) would correspond to the local description. By making the substitution $\mathbf{x} = \mathbf{x}' + \mathbf{v}\,\mathbf{t}'$ (and afterwards dropping the prime) we end up with the substantial description:

$$\frac{\mathrm{d}\bar{\rho}(\mathbf{x}+\mathbf{v}t, \mathbf{v}, t)}{\mathrm{d}t} = \frac{\hat{\lambda}(\mathbf{x}+\mathbf{v}t, \mathrm{t}, \mathbf{v}) - \bar{\rho}(\mathbf{x}+\mathbf{v}t, \mathbf{v}, t)}{T_p}$$

$$-(i/\hbar)[H(\mathbf{x}+\mathbf{v}t, t), \bar{\rho}(\mathbf{x}+\mathbf{v}t, \mathbf{v}, t)] - \tfrac{1}{2}\{\Gamma\rho(\mathbf{x}+\mathbf{v}t, \mathbf{v}, t) + \rho\Gamma\}. \tag{188}$$

When we split the matrix ρ into its elements, we obtain from (188) the eqns (128), (130), (131) (in a specialized form, where N_0 acts as reservoir (compare 2.4) and N_3 is eliminated using the fast decay rate of level 3). Most importantly, $\bar{\rho}_{ik}(\mathbf{x}+\mathbf{v}t, \mathbf{v}, t)$ is replaced by $\rho_{ik,\mu}(t)$. The equations of the atomic motion (188) are thus equivalent.

The equivalence of the corresponding field equations can be shown even more simply.

3.11. SINGLE MODE OPERATION IN GAS LASERS[25]

The method of solution consists again in the iteration procedure described in 3.7. We have to repeat it, however, because the coupling coefficients depend also on time. Making the hypothesis

$$b^*(t) = B^* \exp(i\Omega t), \tag{189}$$

we obtain after the above iteration steps in the same approximation

$$[\kappa + i(\Omega - \omega)]B^* \exp(i\Omega t) = i\sum_{\mu} g_\mu^*(t)\rho_{12,\mu}(t). \tag{190}$$

In this equation, $g_\mu^*(t)$ is the coupling constant defined by (178) and $\rho_{12,\mu}(t)$, in the second approximation, is given by

$$\begin{aligned}\rho_{12,\mu}(t) = {} & B^* \exp(i\Omega t)\{c_+ \exp[i(kx_\mu + vt)] + c_- \exp[-\mathrm{i}(kx_\mu + vt)]\} \\ & + \bar{n}B^* \exp(i\Omega t)\{d_+ \exp[i(kx_\mu + vt)] + d_- \exp[-i(kx_\mu + vt)]\} \\ & + f_+ \exp[3i(kx_\mu + vt)] + f_- \exp[-3i(kx_\mu + vt)] \\ & + j_+ \exp[i(kx_\mu + vt)] + j_- \exp[-i(kx_\mu + vt)],\end{aligned} \tag{191}$$

where $c_\pm$, $d_\pm$, $f_\pm$, $j_\pm$ are complex constants which are independent of space and time, but depend on the mode frequency and on the velocity of the single atom. When we multiply (191) with g_μ^* (see (178)) and sum over the coordinates, the time dependence with $\exp(ivt)$ and $\exp(3ivt)$ drops out on account of the orthogonality properties so that the right hand side has exactly the same time dependence as required for b^* by the ansatz (189).

Integrating over x and averaging over θ explicitly and assuming a Gaussian velocity distribution $w(v)$ as well as a homogeneous spatial distribution of the active atoms, we obtain the following two equations (α and β) for the photon density and frequency up to order γ/α.

α. Equation for the photon density

$$\kappa = \frac{\pi^{\frac{1}{2}} \rho A d_0 \exp -\delta^2}{6\alpha}\left[1 - \frac{3}{5} A\bar{n} T\left\{\frac{1}{2\gamma} + \frac{\gamma}{2(\gamma^2 + (\Omega - v_0)^2)}\right\}\right], \tag{192}$$

where A is defined in (165) and d_0 is the unsaturated inversion of a single atom. From this we can obtain the photon density

$$\bar{n} = \frac{10\gamma}{3AT}\left(1 - \frac{3d_{\text{thr}}(\delta)}{d_0}\right)\left[1 + \frac{\gamma^2}{\gamma^2 + (\Omega - v_0)^2}\right]^{-1}. \tag{193}$$

The photon density shows a dip (known as Lamb dip) if plotted as a function of frequency. It is interesting to note that this dip is brought about by the fact that the atoms move in both directions, so giving rise to two holes being burned into the inhomogeneously broadened line at two symmetric points of the line shape. If we have fixed atoms instead no such dip occurs.

(β) Equation for the frequency shift

$$\Omega - \omega = -\frac{\rho A d_0}{3\alpha} \exp(-\delta^2)\, \Phi(\delta) + \bar{n} C', \tag{194}$$

where A, δ, and $\Phi(\delta)$ are defined in eqns (165), (169) and (167), respectively, and C' is given by

$$C' = \frac{A^2 \rho d_0 T \pi^{\frac{1}{2}}}{2\alpha} \exp(-\delta^2)\left\{\frac{1}{10} \cdot \frac{\Omega - v_0}{\gamma^2 + (\Omega - v_0)^2} + \frac{1}{5}\frac{\delta}{\alpha}\right\}. \tag{195}$$

We compare this result with the expression (166) for fixed atoms and a single direction of polarization. The first sum term on the right-hand side in (194) agrees with the corresponding one in (166) apart from a factor 1/3 which stems from the integration over the polarization (only 1/3 of the atoms

participate in the laser process on the average). This term represents power dependent mode pulling and stems from the Doppler shape of the line.

With respect to the term proportional to $\bar{n}$ which comes from the time independent atomic inversion, we observe that the part of C' which stems from the second sum term in the curly bracket agrees with the total expression C (eqn (166)) apart from a numerical factor which stems from the integration over the polarization angles again. This second term in C' is generally much smaller than the first sum term (provided the detuning is not too strong ($\delta \ll 1$)). It describes the frequency pushing which is due to the existence of two holes burned into the inversion (note, that a standing wave interacts with atoms whose frequencies are shifted both by $+kv$ and $-kv$, where k is the wave number of the mode and v is the velocity of the atom). The frequency pushing becomes dominant if the mode is tuned to the centre of the line within about a natural line width.

Inserting $\bar{n}$ given by eqn (193) into (194) gives, finally

$$\Omega = \omega - \kappa \frac{2}{\pi^{\frac{1}{2}}} \int_0^{\delta} \exp(u^2)\, du + \kappa \left(\frac{d_0}{3 d_{\mathrm{thr}}(\delta)} - 1 \right) \times \left(-\frac{2}{\pi^{\frac{1}{2}}} \int_0^{\delta} \exp(u^2)\, du + \frac{\gamma(\Omega - \nu_0)}{2\gamma^2 + (\Omega - \nu_0)^2} \right). \tag{196}$$

4. The Fully Quantum Mechanical Treatment

4.1. Need for a fully quantum mechanical treatment

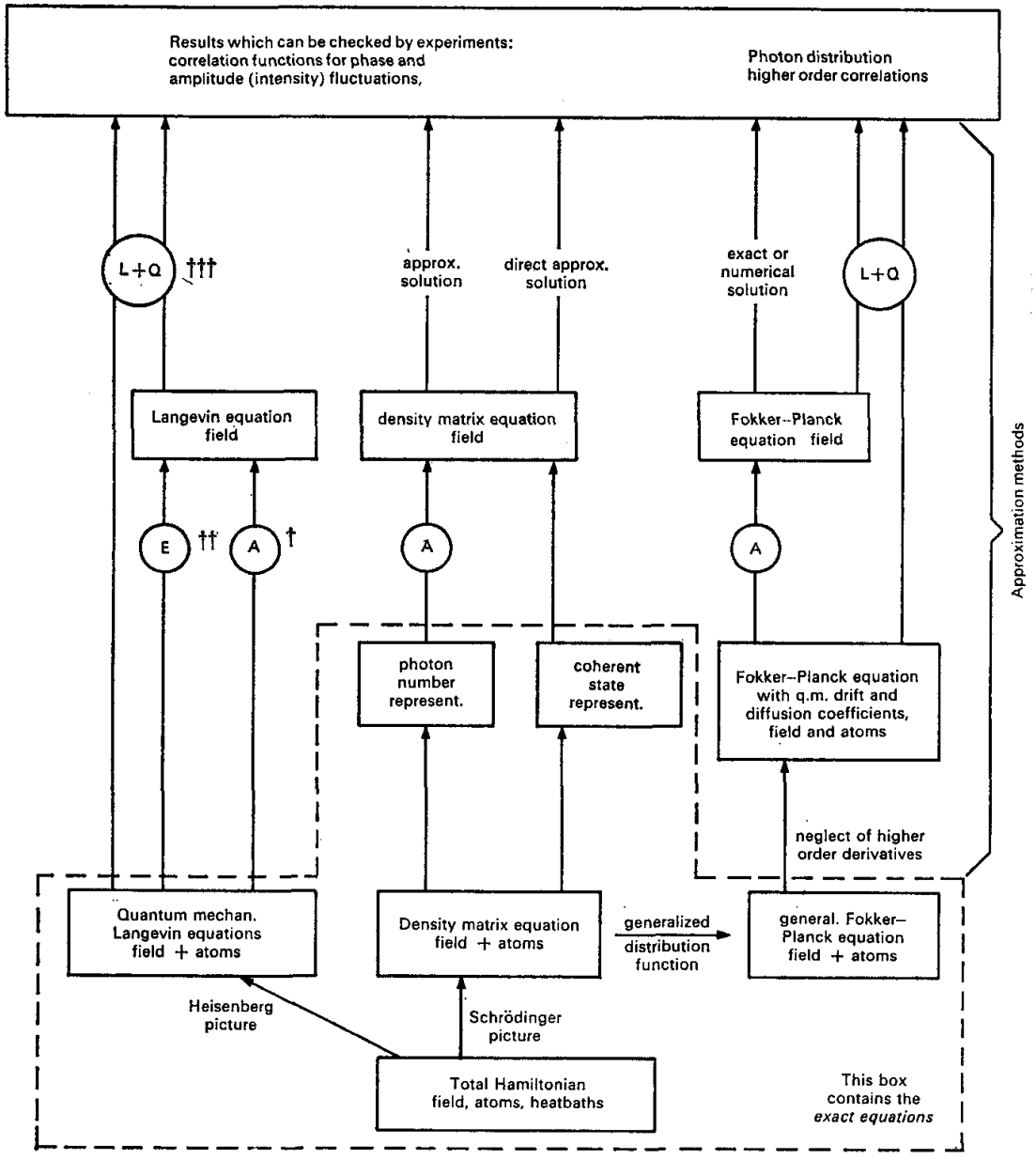
In the foregoing Section 3 we have seen that the semiclassical equations allow us to treat a great number of laser phenomena. These equations have a fundamental drawback, however, they predict that laser action should occur only above a certain threshold, and that the light has an infinitely sharp line (the phase and amplitude are fixed quantities), whereas below that threshold no laser action should occur at all. It is, however, evident, that even below threshold the laser emits light (any ordinary lamp does so) and it is further known that laser light has a finite linewidth above threshold.

Let us discuss the reason for these effects by first considering a region far below threshold, where the laser acts as a usual lamp. Its light stems from spontaneous emission, which manifestly is a *quantum mechanical process*. When the inversion is increased, the spontaneously emitted light paths are enhanced by stimulated emission. This effect has been treated by a great number of authors, using a semi-classical or a quantum mechanical *linear* response theory. They predicted a finite linewidth which decreases with in-

creasing inversion. The field should be a superposition of statistically independent wave tracks so that its amplitude possesses a Gaussian distribution. For quite a while it was rather generally believed that these theories represented the whole story. There was still a puzzle, however—why should one be allowed to use *linear* theories to explain the linewidth, while for all other laser phenomena the *nonlinearity* (treated in the theories of Haken, Sauermann and Lamb) played the decisive role? Thus a theory was needed which was both *quantum mechanical and nonlinear*. In 1964 we developed such a theory.[26] It revealed immediately that the statistical properties of laser light differ basically below and above the laser threshold. Well below threshold the results of the linear treatments were re-obtained. Above threshold, the field amplitude was stable with small quantum mechanical amplitude and phase fluctuations superimposed on it. Speaking more mathematically, it turned out that the creation operator $b_\lambda^\dagger$ of the mode λ could be split into a c-number and a residual operator. The c-number part is exactly the one, which is used in the semiclassical equations. The predictions about the amplitude fluctuations were fully substantiated by experimental results published shortly afterwards.[27]

Our equations can be considered as quantum mechanical analogues of the Langevin equations of classical physics. Besides this kind of laser equation, which have been subsequently used by a number of authors (Sauermann,[28] Lax,[29] Haken,[30] Arzt *et al.*[31]) two further methods were developed, which proved to be more or less equivalent to that approach: the laser master-equation (derived by Weidlich and Haake in 1965[32] (see also Pike and his chapter in this volume; Scully and Lamb,[33] Fleck,[34] Willis,[35] Weidlich *et al.*,[36] Gnutzmann and Weidlich[37]) and the Fokker–Planck treatment of Risken.[38] Risken interpreted our quantum mechanical Langevin equation as a classical one and established its associated classical Fokker–Planck equation. The exact stationary solution allowed him to predict the smooth change of photon statistics from below to above laser threshold. His predictions, as well as further ones on correlation functions, were fully substantiated by experiments.[39] There remained, however, a challenge for the theoreticians working in that field: why is one allowed to use a classical Fokker–Planck equation for a definitely quantum mechanical process? Or in other words: how can one derive a Fokker–Planck equation from a quantum mechanical equation without losing the quantum mechanical information? A first important step in that direction was taken in the paper of Weidlich and Haake. When they chose the density matrix in the coherent state representation (see Glauber's chapter in this volume), they obtained a partial differential equation of the type of a classical Fokker–Planck equation. A further step was taken by Lax and Louisell,[40] who derived a Fokker–Planck equation for the field using a representation equivalent to the *diagonal coherent state* representa-

TABLE II



† A: adiabatic elimination of atomic variables
†† E: exact elimination of atomic variables
††† L+Q: linearization and quantum mechanical quasilinearization

Family tree of the quantum theory of the laser

tion (see Glauber's chapter in this volume). Their treatment of the field is now generally adopted (although there may be some pathological cases where it fails, as shown e.g. by Mollow and Glauber). The laser equations contain, of course, also the atomic variables or, as we will see, the atomic operators. Thus the task remained to derive a Fokker–Planck type equation for those atomic operators. Schmid and Risken[41] proposed a distribution function for a single atom which contained *all* the relevant atomic operators in a symmetrized form, and derived the first two moments for the Fokker–Planck equation. Lax[42] first confined himself to the occupation numbers. Because these quantities commute with each other and with the field operators, this treatment is trivial, however, as stated by Lax and Yuen.[43] Finally, a completely exact generalized Fokker–Planck equation was derived for all noncommuting macroscopic variables of a ground state laser (Haken, *et al.*[44]). If one keeps the leading terms, this equation reduces to one essentially equivalent to that proposed earlier by Risken *et al.*[45] The method of Haken *et al.* was adopted by Lax and Yuen[43] to treat an upper state maser, while Gordon[46] used a distribution function for the atoms of the Schmid–Risken type. Recently, the explicit form of the generalized Fokker–Planck equation for an *arbitrary* dissipative (or non-dissipative) quantum system, provided its density matrix is given, has been derived,[47] so that this problem can be considered as completely solved. This equation is derived in Section 5 of this chapter.

The logical connection between the different approaches is shown in Table II. In view of the numerous approaches which often differ but slightly, one cannot help having the impression that the problem of the quantum theory of the laser has been "overkilled" (as may be the case with many contemporary problems of physics). On the other hand these methods, which allow us to deal with systems far from thermal equilibrium, may have important applications in other fields of physics, e.g. nonlinear quantum optics (parametric optical oscillator, second harmonic generation), critical phenomena of light propagation in continuous media, super-conductivity (e.g. Josephson junctions), nonlinear spinwave theory, tunnel diodes, etc.

In our present treatment we proceed as follows. We first derive the quantum mechanical Langevin equations for a single mode laser and indicate their solution and the physical consequences. We then sketch the derivation of the laser master equation, derive the exact generalized Fokker–Planck equation and discuss the approximations which reduce it eventually to Risken's original equation. Finally we discuss its numerical solution and compare it with experiments. To readers who are not so much interested in the sophisticated quantum mechanical derivation of the Fokker–Planck equation we offer an alternative derivation in the framework of the purely classical theory (4.12.c).

4.2. THE HAMILTONIAN AND HEISENBERG EQUATIONS OF MOTION FOR THE LOSS-LESS SYSTEM

To avoid unnecessary complications, we treat a single mode represented by a running wave, and a system of atoms with two levels and a homogeneously broadened line. Occasionally we shall indicate the changes involved when several modes are present, or when three-level atoms are used, or when the line is inhomogeneously broadened.

The Hamiltonian of the total system (field and atoms) may be derived by the standard methods of quantum-electrodynamics (see for example, the chapter by Kibble in this volume).

The field

The vector potential $\mathbf{A}(\mathbf{x})$ of the electromagnetic field is decomposed into the "cavity" modes $\mathbf{U}_\lambda(\mathbf{x})$ thus,

$$\mathbf{A}(\mathbf{x}) = \sum_\lambda \left(\frac{2\pi c^2 \hbar}{\omega_\lambda}\right)^{\frac{1}{2}} (b_\lambda + b_\lambda^\dagger)\mathbf{U}_\lambda(\mathbf{x}), \tag{197}$$

where ω_λ is the mode frequency in the "unloaded" cavity and $b_\lambda^\dagger$ and b_λ are the creation and annihilation operators of the mode λ. These operators obey the commutation relations

$$\begin{aligned} b_\lambda b_{\lambda'}^\dagger - b_{\lambda'}^\dagger b_\lambda &= \delta_{\lambda\lambda'}, \\ b_\lambda b_{\lambda'} - b_{\lambda'} b_\lambda &= 0, \\ b_\lambda^\dagger b_{\lambda'}^\dagger - b_{\lambda'}^\dagger b_\lambda^\dagger &= 0. \end{aligned} \tag{198}$$

The Hamiltonian of the field is given by

$$H_f = \sum_\lambda \hbar\omega_\lambda b_\lambda^\dagger b_\lambda. \tag{199}$$

In the following we retain only a single mode and drop the index λ. The Hamiltonian then reads

$$H_f = \hbar\omega b^\dagger b. \tag{200}$$

The matter

In accordance with the foregoing sections, we consider a set of laser-active atoms, which we distinguish by an index μ and which are embedded in a solid state matrix at random sites, $\mathbf{x}_\mu$. We treat explicitly only a single electron ("Leucht-electron") of each atom. The electrons are described by the formalism of second quantization. The creation and annihilation operators of the

electrons of the atom μ in the state j (= 0, 1, 2, ...) are, respectively, $a_{\mu j}^{\dagger}$ and $a_{\mu j}$. These operators obey the anticommutation relations

$$\begin{aligned} a_{\mu j}^{\dagger} a_{\mu' j'} + a_{\mu' j'} a_{\mu j}^{\dagger} &= \delta_{\mu\mu'} \delta_{jj'}, \\ a_{\mu j} a_{\mu' j'} + a_{\mu' j'} a_{\mu j} &= 0 \\ a_{\mu j}^{\dagger} a_{\mu' j'}^{\dagger} + a_{\mu' j'}^{\dagger} a_{\mu j}^{\dagger} &= 0. \end{aligned} \tag{201}$$

The Hamiltonian of the total atomic system reads

$$H_a = \sum_{\mu j} E_{\mu j} a_{\mu j}^{\dagger} a_{\mu j}, \tag{202}$$

where $E_{\mu j}$ is the energy of the level j of the atom μ. In the following we treat mainly two-level atoms with a homogeneously broadened line. Choosing $E_{\mu 1} = 0$ and putting $E_{\mu 2} = \hbar\nu$, we obtain

$$H_a = \hbar\nu \sum_{\mu} a_{\mu 2}^{\dagger} a_{\mu 2}. \tag{203}$$

The interaction Hamiltonian

The Hamiltonian describing the interaction between a set of laser modes λ and the atomic system μ is given by

$$H_i = \hbar \sum_{\lambda\mu} \{(a_2^{\dagger} a_1)_{\mu} b_{\lambda} g_{\mu\lambda}^{*} + (a_1^{\dagger} a_2)_{\mu} b_{\lambda}^{\dagger} g_{\mu\lambda}\}, \tag{204}$$

where

$$(a_2^{\dagger} a_1)_{\mu} = a_{\mu 2}^{\dagger} a_{\mu 1}, \text{ etc.}$$

The terms $(a_2^{\dagger} a_1)_{\mu} b_{\lambda}$ describe the excitation of the atom μ connected with the absorption of a photon of the mode λ, whereas $(a_1^{\dagger} a_2)_{\mu} b_{\lambda}^{\dagger}$ describes the inverse process. To obtain H_i from usual quantum electrodynamics we made two approximations:

1. we neglected terms quadratic in **A** (i.e. terms quadratic or bilinear in $b_{\lambda}, b_{\lambda}^{\dagger}$);
2. we neglected antiresonant terms of the form $a_2^{\dagger} a_1^{\dagger} b^{\dagger}$, which describe the simultaneous electronic excitation and creation of a photon and $a_1^{\dagger} a_2 b$, which describe the simultaneous electronic recombination and annihilation of a photon.

The coupling constants $g_{\mu\lambda}$ are in the so-called dipole approximation identical with the ones given in eqn (121). When we consider only a single mode, we write

$$H_i = \hbar\left\{b\sum_{\mu}(a_2^{\dagger}a_1)_{\mu}g_{\mu}^{*}+b^{\dagger}\sum_{\mu}(a_1^{\dagger}a_2)_{\mu}g_{\mu}\right\}. \tag{205}$$

The total Hamiltonian

This is given by

$$H = H_f+H_a+H_i. \tag{206}$$

The equations of motion

For the laser theory it proved most fruitful to treat the Heisenberg operator equations, because they exhibit a close correspondence to the semiclassical equations (122)–(124). Using the relation

$$\frac{\mathrm{d}\Omega}{\mathrm{d}t} = \frac{i}{\hbar}[H,\Omega] \tag{207}$$

for the time dependence of an arbitrary operator Ω, we find for the creation operator $b_{\lambda}^{\dagger}$ the equation

$$b_{\lambda}^{\dagger} = i\omega_{\lambda}b_{\lambda}^{\dagger}+i\sum_{\mu}g_{\mu\lambda}^{*}(a_2^{\dagger}a_1)_{\mu}. \tag{208}$$

With respect to the electronic operators $a_{\mu j}^{\dagger}$, $a_{\mu j}$, it turned out advantageous to derive equations of motion for pairs $(a_j^{\dagger}a_k)_{\mu}$ rather than for the single operators $a_{\mu j}^{\dagger}$ or $a_{\mu j}$. We obtained

$$(a_2^{\dagger}a_1)^{\cdot}_{\mu} = iv_{\mu}(a_2^{\dagger}a_1)_{\mu}-i\sum_{\mu}g_{\mu\lambda}b_{\lambda}^{\dagger}(a_2^{\dagger}a_2-a_1^{\dagger}a_1)_{\mu} \tag{209}$$

and a corresponding equation for the hermitian conjugate of $(a_2^{\dagger}a_1)_{\mu}$. Because the operator $(a_2^{\dagger}a_2-a_1^{\dagger}a_1)_{\mu}$, which describes the atomic inversion, occurs in eqn (209), we have to derive a further equation for it. By letting

$$\sigma_{\mu} = (a_2^{\dagger}a_2-a_1^{\dagger}a_1)_{\mu}, \tag{210}$$

we obtain

$$\dot{\sigma}_{\mu} = 2i\sum_{\lambda}(g_{\mu\lambda}(a_1^{\dagger}a_2)_{\mu}b_{\lambda}^{\dagger}-g_{\mu\lambda}^{*}(a_2^{\dagger}a_1)_{\mu}b_{\lambda}). \tag{211}$$

4.3. THE HEISENBERG EQUATIONS OF MOTION INCLUDING DAMPING

The quantum mechanical Langevin equations

To compare these quantum mechanical equations with the semiclassical ones (122), (123) and (124), we note that by making the following changes

$$b^\dagger \to b^*; \quad a_2^\dagger a_1 \to \rho_{12}; \quad \sigma_\mu \to d_\mu, \tag{212}$$

we see immediately that both sets of equations are identical apart from one basic difference, viz. the semiclassical equations contain damping terms as follows

(122): $$-\kappa b^* \tag{213}$$

(123): $$-\gamma \rho_{12} \tag{214}$$

(124): $$\frac{d_{0\mu} - d_\mu}{T} \tag{215}$$

These terms are not in the quantum mechanical equations (208), (209) and (211). In Section 3 we have seen, that just these damping constants determine qualitatively and even quantitatively the laser phenomena, threshold, intensity, actual frequency, etc. Thus the terms (213), (214) and (215) must be incorporated into eqns (208), (209) and (211). Let us try to do this first in eqn (208), where we put $g = 0$ for the time being. The solution of the equation

$$\dot{b}^\dagger = (i\omega - \kappa)\, b^\dagger \tag{216}$$

is

$$b^\dagger = b^\dagger(0) \exp[(i\omega - \kappa)\, t]. \tag{217}$$

Inserting this into one of the commutation relations (198) we find that

$$[b, b^\dagger] = \exp -2\kappa t, \tag{218}$$

i.e. the commutation relations (198) are violated.

The reason for the violation of the commutation relations can be found as follows. The damping of the field is caused by other heatbaths or reservoirs. For instance, the losses in the mirrors stem from the interaction of the light field with the electrons in the mirrors. Thus, in order to describe the damping properly, we must include these reservoirs in the Hamiltonian from the very beginning. When we now try to eliminate these heatbaths, it turns out that they lead not only to damping but also to additional fluctuations. These

fluctuations act as random forces on the field operator $b^\dagger$, so that the quantum mechanically consistent equation must read

$$\dot{b}^\dagger = (i\omega - \kappa)\, b^\dagger + F^\dagger(t) \tag{219}$$

instead of (208).

The forces $F^\dagger(t)$, $F(t)$ derived by Senitzky[48] are non-commuting operators which have quite general properties which do not depend on the special heatbath used. In the appendix we show how the heatbath co-ordinates can be eliminated and how these quantum mechanical Langevin forces arise. In fact the analogy with a classical Brownian motion is quite close. The fluctuating forces F are of fundamental importance for the determination of the phase- and amplitude-fluctuations of the maser (microwave region), because they represent *thermal* noise. In the optical region the *spontaneous emission noise* plays the decisive role. This effect was first considered by Haken[26] in the following way. The eqns (209) and (211) alone are not sufficient to treat spontaneous emission by the Wigner–Weisskopf method, but must be supplemented by the initial condition that the atom is in its upper state. In the case of the ammonia maser this "preparation" of the atoms is done when they are shot into the cavity at random times t_ν. In the case of fixed atoms the preparation of the initial state is achieved by the pumping process. In a completely symmetric way the initial state is also prepared, for example, by the spontaneous emission into the nonlasing modes (which form a continuum). Because eqn (209) is a first order differential equation, such initial conditions can be built in by adding on the right hand side a term

$$\Gamma_{\mu,21}(t) = \sum_\nu \delta(t - t_{\mu\nu})\{(a_2^\dagger a_1)_\mu(t_{\mu\nu}+0) - (a_2^\dagger a_1)_\mu(t_{\mu\nu}-0)\}, \tag{220}$$

where, for example, $(a_2^\dagger a_1)_\mu\,(t_{\mu\nu}+0)$ is the "value" of the operator immediately after the pumping process. One may show that the Γ's are non-commuting operators with the property

$$\langle \Gamma_\mu \rangle = 0, \quad \langle \Gamma_{\mu,i_1 i_2}(t_1)\Gamma_{\mu,j_1 j_2}(t_2)\rangle = G_{i_1 i_2, j_1 j_2}\,\delta(t_1 - t_2), \tag{221}$$

where the δ-function expresses the fact that the external perturbations ("heatbaths" or "reservoirs") which "prepare" the initial state have a short memory (compared to the other time-constants in the system), or in other words, that they are "Markoffian".

Another, more rigorous, way is to couple the electrons of the single atoms to heatbaths which represent the pumping, the spontaneous decay into nonlasing modes, interaction with lattice vibrations, collisions etc. and to eliminate then these heatbath variables. This has been done for a two-level system in detail by Sauermann.[49] Another method to derive the properties of the

coefficients G in equation (221) which was used by Haken and Weidlich[50] relies on the requirement of quantum mechanical consistence. Summarizing their method, we first consider equations for the occupation number average,

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle a_2^\dagger a_2\rangle = w_{12}\langle a_1^\dagger a_1\rangle - w_{21}\langle a_2^\dagger a_2\rangle, \tag{222}$$

and the average for the dipole moment operators

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle a_2^\dagger a_1\rangle = (i\nu-\gamma)\langle a_2^\dagger a_1\rangle, \tag{223}$$

for which we may write down phenomenological equations in the sense of the semiclassical treatment, where w_{12}, w_{21} are the atomic transition rates, ν is the circular frequency of the atomic transitions, and γ is the atomic half-width. The quantities

$$a_i^\dagger a_k = P_{ik} \tag{224}$$

are projection operators which project from one state to another and which obey the following relation for projection operators in the single electron space

$$P_{ik} P_{lm} = P_{im}\delta_{kl}. \tag{225}$$

This relation is violated if the operators are forced to obey equations with damping constants. In order to save the property (225), we introduce fluctuating forces $\Gamma(t)$ with the property (221) so that our fully quantum mechanical equations now read for the operators of the two level system:

$$\mathrm{d}/\mathrm{d}t\,(a_2^\dagger a_2) = w_{12}\, a_1^\dagger a_1 - w_{21}\, a_2^\dagger a_2 + \Gamma_{22}(t); \tag{226}$$

$$\mathrm{d}/\mathrm{d}t\,(a_1^\dagger a_1) = -w_{12}\, a_1^\dagger a_1 + w_{21}\, a_2^\dagger a_2 + \Gamma_{11}(t); \tag{227}$$

$$\mathrm{d}/\mathrm{d}t\,(a_2^\dagger a_1) = (i\nu-\gamma)\, a_2^\dagger a_1 + \Gamma_{21}(t). \tag{228}$$

The properties of the fluctuating forces Γ are derived in Section 5 for an arbitrary quantum system coupled to Markoffian heatbaths. We quote a few examples:

$$G_{12,\,21} = 2\gamma(\langle a_1^\dagger a_1\rangle + \langle a_2^\dagger a_2\rangle)$$

$$G_{12,\,12} = 0, \qquad G_{21,\,21} = 0.$$

In particular it turns out that the relations remain unaltered, if the action of external fields is admitted. Thus we can combine the form of eqns (226), (227) and (228) with those containing the coherent interaction:

$$\dot{b}_\lambda^\dagger = (i\omega_\lambda - \kappa_\lambda) b_\lambda^\dagger + i \sum_\mu g_{\mu\lambda}^* (a_2^\dagger a_1)_\mu + F_\lambda^\dagger(t) \tag{229}$$

$$(a_2^\dagger a_1)_\mu^\cdot = (iv_\mu - \gamma)(a_2^\dagger a_1)_\mu - i \sum_\lambda g_{\mu\lambda} b_\lambda^\dagger (a_2^\dagger a_2 - a_1^\dagger a_1)_\mu + \Gamma_{21,\mu}(t) \tag{230}$$

$$\dot{\sigma}_\mu = \frac{d_0 - \sigma_\mu}{T} + 2i \sum_\lambda \left(g_{\mu\lambda} (a_1^\dagger a_2)_\mu b_\lambda^\dagger - g_{\mu\lambda}^* (a_2^\dagger a_1)_\mu b_\lambda \right) + \Gamma_{\sigma\mu} \tag{231}$$

where

$$d_0 = \frac{w_{21} - w_{12}}{w_{12} + w_{21}}; \quad \frac{1}{T} = w_{12} + w_{21}; \quad \Gamma_\sigma = \Gamma_{22} - \Gamma_{11}.$$

4.4. THE QUANTUM MECHANICAL LANGEVIN EQUATIONS FOR A SET OF FIELD MODES AND A SYSTEM OF N LEVEL ATOMS. CONNECTION WITH THE SEMICLASSICAL EQUATIONS†

The methods given in Section 5 enable quantum mechanically consistent equations for an arbitrary quantum system to be derived. In the case of the laser, the corresponding equations are:

a. Quantum mechanical equations

α. The field equations

$$\frac{\mathrm{d}}{\mathrm{d}t} b_\lambda^\dagger = (i\omega_\lambda - \kappa_\lambda) b_\lambda^\dagger + i \sum_{jl\mu} g_{jl\mu\lambda} (a_j^\dagger a_l)_\mu + F_\lambda^\dagger(t), \tag{232}$$

where $b_\lambda^\dagger$ is the creation operator of the cavity mode λ, ω_λ its frequency, and $t_\lambda = \frac{1}{2}\kappa_\lambda$ its lifetime (both in the unloaded cavity). $(a_j^\dagger a_l)_\mu$ is an abbreviation for $a_{j\mu}^\dagger a_{l\mu}$ where $a_{j\mu}^\dagger$ is the creation operator of an electron in the level j at an atom μ (μ denotes, for instance, the lattice site) and $a_{l\mu}$ is correspondingly the annihilation operator. $g_{jl\mu\lambda}$ describes the coupling of the mode λ with the electron at atom μ, which causes a real or virtual transition between levels l and j. The fluctuating forces $F_\lambda(t)$ are characterized by the properties

$$\langle F_\lambda(t) F_{\lambda'}(t') \rangle = 2\kappa_\lambda n_\lambda(T) \delta(t-t') \delta_{\lambda\lambda'}$$

$$\langle F_\lambda(t) F_{\lambda'}^\dagger(t') \rangle = 2\kappa_\lambda (n_\lambda(T)+1) \delta(t-t') \delta_{\lambda\lambda'}$$

$$\langle F_\lambda F_{\lambda'} \rangle = \langle F_\lambda^\dagger F_{\lambda'}^\dagger \rangle = \langle F_\lambda \rangle = \langle F_\lambda^\dagger \rangle = 0.$$

$n_\lambda(T)$ is the number of thermal quanta at the temperature T.

† The reading of this section is not required for the subsequent ones.

(β) The matter equations

$$\frac{\mathrm{d}}{\mathrm{d}t}(a_{j'}^{\dagger}\, a_{l'})_{\mu} = \nu_{j'l'\mu}\,(a_{j'}^{\dagger}\, a_{l'})_{\mu} + i\sum_{jl\lambda} g_{jl\mu\lambda}\,(b_{\lambda}^{\dagger}+b_{\lambda})\{(a_{j}^{\dagger}\, a_{l'})_{\mu}\,\delta_{lj'} - (a_{j'}^{\dagger}\, a_{l})_{\mu}\,\delta_{jl'}\}$$
$$-\gamma_{j'l'}(a_{j'}^{\dagger}\, a_{l'})_{\mu}\,(1-\delta_{j'l'}) + \delta_{j'l'}\left\{\sum_{k}(w_{kj'}\,(a_{k}^{\dagger}\, a_{k})_{\mu}\right.$$
$$\left.-(a_{j'}^{\dagger}\, a_{j'})_{\mu}\, w_{j'k})\right\} + \Gamma_{j'l'\mu}(t) \qquad (233)$$
$$\nu_{jl\mu} = \frac{1}{\hbar}(E_j - E_l)_{\mu}.$$

E_j is the energy of an electron in the level j at atom μ. The inhomogeneous broadening is taken into account by letting the E's explicitly depend on μ. δ_{lj} is Kronecker's symbol ($= 1$ for $l = j$; $= 0$ for $l \neq j$). The Γ's are the fluctuating forces. It is assumed that they are uncorrelated for different μ's and have the properties (221) where the coefficients in (221) are given by (490). *The semiclassical equations.* If we take the average with respect to the heatbath variables which still occur in the F's and Γ's, the fluctuating forces vanish. We take further traces with respect to the electronic states and the light field and make the approximation that[51]

$$\mathrm{tr}_{\mathrm{total}}(b_{\lambda}\, a_{l}^{\dagger}\, a_{j}\, \rho_{\mathrm{tot}}) = \mathrm{tr}_{\mathrm{field}}(b_{\lambda}\, \rho_{\mathrm{field}})\,\mathrm{tr}_{\mathrm{el}}(a_{l}^{\dagger}\, a_{j}\, \rho_{\mathrm{atom}}). \qquad (234)$$

ρ_{tot}, ρ_{field} and ρ_{atom} are the density matrices of the total system, of the field and of the atoms, respectively. Then, using

$$\mathrm{tr}_{\mathrm{el}}(a_{l}^{\dagger}\, a_{j}\, \rho_{\mathrm{atom}})_{\mu} = \rho_{jl\mu}, \mathrm{tr}_{\mathrm{field}}(b_{\lambda}\, \rho_{\mathrm{field}}) = \langle b_{\lambda}\rangle, \qquad (235)$$

we obtain finally from (232) and (233) the so-called "semiclassical" density matrix equations.

b. "Semiclassical" density matrix equations[52]

When we specialize the above equations to the four level case and apply the rotating wave approximation, we obtain the equations (127)–(132) with

$$g_{12\mu\lambda} = g_{\mu\lambda}; \qquad g_{21\mu\lambda} = g_{\mu\lambda}^{*}.$$

4.5 QUALITATIVE DISCUSSION OF SINGLE MODE OPERATION[26,53]

Some transformations

A series of implications of the above equations can be explained by the following specialization. We consider a system of two-level atoms with a single transition frequency ν, which are in resonance with a single, running

cavity mode. The equations of motion then are

$$\dot{b}^\dagger = (i\omega - \kappa)\, b^\dagger + i \sum_\mu g \exp(-ikx_\mu)\, (a_2^\dagger a_1)_\mu + F^\dagger, \tag{236}$$

$$(a_2^\dagger a_1)_\mu^\cdot = (i\omega - \gamma)(a_2^\dagger a_1)_\mu - ig \exp(ikx_\mu)\, b^\dagger (a_2^\dagger a_2 - a_1^\dagger a_1)_\mu + \Gamma_{21,\mu}, \tag{237}$$

together with the complex conjugate equation, and further

$$(a_2^\dagger a_2)_\mu^\cdot = (a_1^\dagger a_1)_\mu w_{12} - (a_2^\dagger a_2)_\mu w_{21} + i(a_1^\dagger a_2)_\mu g_\mu b^\dagger - i(a_2^\dagger a_1)_\mu g_\mu^* b + \Gamma_{22,\mu}, \tag{238}$$

$$(a_1^\dagger a_1)_\mu^\cdot = (a_2^\dagger a_2)_\mu w_{21} - (a_1^\dagger a_1)_\mu w_{12} - i(a_1^\dagger a_2)_\mu g_\mu b^\dagger + i(a_2^\dagger a_1)_\mu g_\mu^* b + \Gamma_{11,\mu}. \tag{239}$$

We eliminate ω and the factors $\exp(ikx_\mu)$ and $\exp(-ikx_\mu)$ from the above equations by the transformations

$$b^\dagger \to b^\dagger \exp(i\omega t), \qquad b \to b \exp(i\omega t), \tag{240}$$

$$\begin{aligned} (a_2^\dagger a_1)_\mu &\to \exp(ikx_\mu) \exp(i\omega t)\, (a_2^\dagger a_1)_\mu, \\ (a_1^\dagger a_2)_\mu &\to \exp(-ikx_\mu) \exp(-i\omega t)\, (a_1^\dagger a_2)_\mu. \end{aligned} \tag{241}$$

We then put $F^\dagger = \tilde{F}^\dagger \exp(i\omega t)$ and $\Gamma_{21} = \exp(i\omega t)\, \tilde{\Gamma}_{21}$. Because the equation $a_1^\dagger a_1 + a_2^\dagger a_2 = 1$ holds exactly, we introduce the new variables

$$a_1^\dagger a_1 + a_2^\dagger a_2 \quad \text{and} \quad (a_2^\dagger a_2 - a_1^\dagger a_1) = \sigma_\mu. \tag{242}$$

The eqns (236)–(239) then take the very simple form

$$\dot{b}^\dagger = -\kappa b + ig \sum_\mu (a_2^\dagger a_1)_\mu + \tilde{F}^\dagger, \tag{243}$$

$$(a_2^\dagger a_1)_\mu^\cdot = -\gamma (a_2^\dagger a_1)_\mu - ig b^\dagger \sigma_\mu + \tilde{\Gamma}_{21,\mu}, \tag{244}$$

$$\dot{\sigma}_\mu = \frac{d_0 - \sigma_\mu}{T} + 2ig(a_1^\dagger a_2\, b^\dagger - a_2^\dagger a_1\, b)_\mu + (\Gamma_{22} - \Gamma_{11})_\mu, \tag{245}$$

where

$$d_0 = \frac{w_{12} - w_{21}}{w_{12} + w_{21}}, \tag{246}$$

and

$$(1/T) = (w_{12} + w_{21}) \tag{247}$$

and T is the relaxation time of the atomic inversion.

Because the mode interacts with the atoms through a sum, we are led to introduce $\sum_\mu (a_2^\dagger a_1)_\mu$ as a new variable $S^\dagger$ and correspondingly to put

$$S = \sum_\mu \sigma_\mu. \tag{248}$$

The eqns (243)–(245) thus simplify (by summation over μ in (244) and (245)) to give

$$\dot{b}^\dagger = -\kappa b^\dagger + igS^\dagger + \tilde{F}^\dagger, \tag{249}$$

$$\dot{S}^\dagger = -\gamma S - igb^\dagger S + \tilde{\Gamma}_{21}, \tag{250}$$

where

$$\tilde{\Gamma}_{21} = \sum_\mu \tilde{\Gamma}_{21,\mu},$$

$$\dot{S} = \frac{S_0 - S}{T} + 2ig(S^- b^\dagger - bS^+) + \Gamma_{22} - \Gamma_{11}, \tag{251}$$

$$\Gamma_{jj} = \sum_\mu \Gamma_{jj,\mu}, \qquad S_0 = Nd_0. \tag{252}$$

and N is the total number of atoms. The variable S^+ can be visualized as the macroscopic, total dipole moment of all atoms, while $S = (N_2 - N_1)$ is the total occupation difference (total inversion). S^+, S^- and S can also be interpreted as operators for the total spin

$$S = (S_x, S_y, S_z),$$

where

$$S^+ = S_x + iS_y, \quad S^- = S_x - iS_y, \quad S = 2S_z.$$

From the first two equations we eliminate S^+ and we substitute S^+ in eqn (251) by means of eqn (249) and S^- by means of an equation which is the complex conjugate of eqn (249). This leaves us with

(248), (249)
$$\ddot{b}^\dagger + (\kappa + \gamma)\dot{b}^\dagger + (\kappa\gamma - g^2 S) b^\dagger = ig\tilde{\Gamma}_{21} + \dot{\tilde{F}}^\dagger + \gamma\tilde{F}^\dagger, \tag{253}$$

where we substitute

$$ig\tilde{\Gamma}_{21} + \dot{\tilde{F}}^\dagger + \gamma\tilde{F}^\dagger = \tilde{F}_{\text{tot}}^\dagger(t),$$

(250), (248)
$$\dot{S} = \frac{S_0 - S}{T} - 2\big((b^\dagger b)^\cdot + 2\kappa b^\dagger b - (b^\dagger \tilde{F} + \tilde{F}^\dagger b)\big) + \Gamma_{22} - \Gamma_{11}. \tag{254}$$

a. The linear range (subthreshold region)

For an interpretation of eqns (253) and (254) we treat $b^\dagger$ as a classical, complex, time dependent variable, which we decompose into its real and imaginary parts

$$b^\dagger(t) = x(t) + iy(t). \tag{255}$$

Equation (253) can then also be decomposed into a real and an imaginary part. The resulting equations then have the same appearance as those of a classical particle, moving in two dimensions under a force

$$\hat{K}_x = -(\kappa\gamma - g^2 S(x,y))\, x, \qquad \hat{K}_y = -(\kappa\gamma - g^2 S(x,y))\, y.$$

Although the motion of the light mode has, of course, nothing to do with a particle motion, this formal analogy is most useful for an understanding of the nonlinear eqns (253) and (254). Consider first the case, that the inversion $S = (N_2 - N_1)$ is kept fixed, which is actually done in the linear theories of laser noise, or in the amplifier range. Let us start with a small inversion, so that

$$\kappa\gamma - g^2 S > 0. \tag{256}$$

Equation (253) is then that of a damped harmonic oscillator, subjected to a random force $K(t)$. This random force can be interpreted in such a way, that it exerts pushes at a random time sequence. After each such excitation the "particle falls down the potential curve" (compare Fig. 4). When we increase

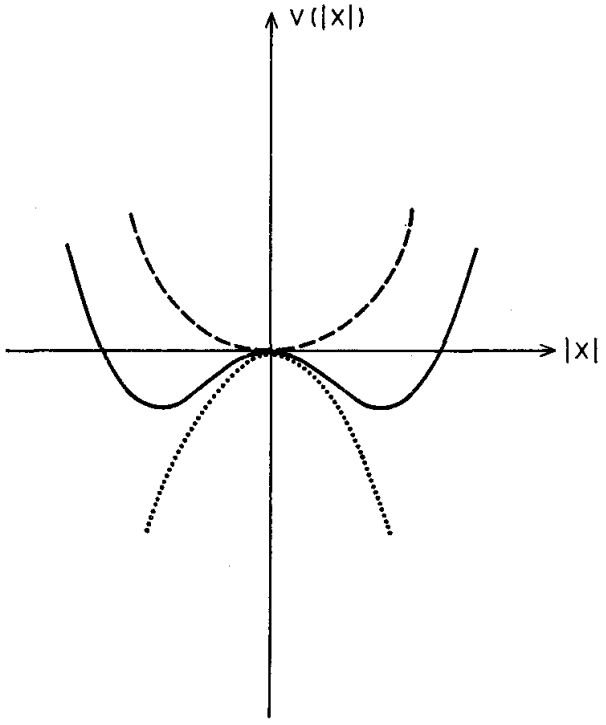


FIG. 4

the inversion S, $(\kappa\gamma - g^2 S)$ becomes smaller so that the particle comes down the potential curve more slowly. Translating this statement into the language of waves, we may say, that the modulation frequency, caused by the fluctuations, becomes smaller, so that a line narrowing results. The pushes in $\tilde{F}_{\text{tot}}(t)$

stem from statistically independent events. Consider for instance Γ_{21} which is equal the sum of the atomic noise operators $\Gamma_{21,\mu}$. Each operator is responsible for the fluctuations of the atomic dipole moment, which are indeed statistically independent. As we will see later explicitly, Γ_{21} is responsible for the spontaneous emission noise, while F accounts for the thermal noise of the walls and the vacuum fluctuations. Each emission process creates a contribution to $b^\dagger$. Due to the flatter curve the amplitude falls down more slowly, or in other words, the losses are partly compensated by induced emission. Because the total amplitude $b^\dagger$ is a sum of many statistical contributions, the probability P of finding a definite value of $b^\dagger$ is a Gaussian,

$$P(b) = \text{const} \exp(-|b|^2/|b_0|^2). \tag{257}$$

When S becomes so big, that $\kappa\gamma - g^2 S > 0$, the system *seems* to become unstable (compare Fig. 4). In the linear theories, S can never become bigger than $S_c = \kappa\gamma/g^2$, because otherwise the light output $\sim|b|^2$ would become infinite. These latter considerations are, however, incorrect, because they neglect the dependence of S on $b^\dagger$ completely.

b. The nonlinear range (at threshold and somewhat above)

Our discussion is based on eqns (253) and (254) where we investigate now the consequence of the dependence of S on b. On account of the line narrowing we may expect that the frequencies inherent in $(b^\dagger b)$ are smaller than κ so that we neglect $(b^\dagger b)^\cdot$ compared with $2\kappa b^\dagger b$. Because the atomic relaxation time T is also small compared to times inherent in $(b^\dagger b)$ (again on account of the line narrowing) S can follow the motion of $b^\dagger b$ adiabatically. Neglecting the fluctuating terms in (254) we thus obtain $S \approx S_0 - 4\kappa T b^\dagger b$ so that (253) takes now the form

$$\ddot{b}^\dagger + (\kappa+\gamma)\dot{b}^\dagger + (\kappa\gamma - g^2 S_0) b^\dagger + 4g^2 \kappa T b^\dagger b b^\dagger = \tilde{F}_{\text{tot}}(t), \tag{258}$$

which can be understood as the equation of a particle moving in the potential

$$V(|b|) = (\kappa\gamma - g^2 S_0)\frac{|b|^2}{2} + g^2 \kappa T |b|^4. \tag{259}$$

For $\kappa\gamma - g^2 S > 0$, i.e. small inversion, the potential curve has qualitatively the same appearance as discussed under (a). For $\kappa\gamma - g^2 S < 0$ the amplitude oscillates around a new stable value $r_0 \neq 0$. Because the potential has rotation symmetry, there is no restoring force in a tangential direction. Thus the phase undergoes some kind of diffusion under the action of the random force $\tilde{F}_{\text{tot}}(t)$. We write

$$b^\dagger(t) = \exp[i\phi(t)]\,(r_0 + \rho(t)) \tag{260}$$

where r_0 is the stable value of the mode amplitude and $\phi(t)$ is the fluctuating

phase of the mode, and discuss the phase diffusion and the amplitude fluctuation ρ in more detail.

α *Phase diffusion.* With increasing S the stable value r_0, of the mode amplitude increases. Consequently the path of the particle, and thus its migration time, increases. Because in a diffusion process we have $\Delta x^2 \propto t$, and for a tangential motion $\Delta x = r_0 \Delta\phi$ we have

$$\overline{(\Delta\phi)^2} \propto t/r_0{}^2. \tag{261}$$

The factor of t (time), however, is just the linewidth. With increasing inversion, the linewidth $\Delta\nu$ due to phase diffusion is proportional to $1/r_0{}^2$ and, because the coherent output power, P, is $\hbar\omega 2\kappa r_0{}^2$, we see that

$$\Delta\nu \propto 1/P. \tag{262}$$

β *Amplitude (intensity) fluctuations.* With increasing r_0, the slope of the potential curve becomes steeper in the vicinity of r_0. Therefore:

1. the oscillation frequency increases, so that the relaxation time of the amplitude fluctuation decreases and $\tau \propto 1/P$;
2. the fluctuating part, ρ, of the amplitude decreases, so

$$\langle \rho^2 \rangle \propto 1/P. \tag{263}$$

c. The nonlinear range at high inversion

With increasing inversion the oscillation frequency of the amplitude fluctuations becomes finally bigger than 2κ, so that the dominant terms in (254) are given by

$$\dot{S} \propto \frac{S_0 - S}{T} - 2(b^\dagger b)^\cdot \tag{264}$$

(the terms with F and $\Gamma_{22} - \Gamma_{11}$ are not completely negligible, however) which, for rapid oscillation has the solution

$$S = \text{const} - 2b^\dagger b. \tag{265}$$

Thus the behaviour is qualitatively similar to that of the region (b).

d. Exact elimination of all atomic coordinates

For the qualitative discussion we have expressed S only approximately by means of $b^\dagger$, b. However, this can be done exactly,

$$S = d_0 - 2\int^t \exp\left(-\frac{1}{T}(t-\tau)\right)\{(b^\dagger b)^\cdot + 2\kappa(b^\dagger b) - (b^\dagger F + F^\dagger b) - \tfrac{1}{2}(\Gamma_{22} - \Gamma_{11})\}\, d\tau. \tag{266}$$

Inserting (266) into (253) yields an exact equation for the b-field alone.

e. The quantum mechanical van der Pol oscillator

As we have just discussed, in a region not too far away from laser threshold, the actual relaxation times of the field are smaller than the decay constants κ and γ. This allows us to neglect the term $\ddot{b}^\dagger$ compared to $(\kappa + \gamma)\,\dot{b}^\dagger$ in eqn (253). Inserting $S = S_0 - 4\kappa T b^\dagger b$ into (253), and neglecting the cavity half-width κ compared to the atomic half-width γ we obtain the equation

$$\dot{b}^\dagger + (\kappa - \frac{g^2}{\gamma} S_0 + \frac{g^2}{\gamma} 4\kappa T b^\dagger b) b^\dagger = \frac{1}{\gamma} \tilde{F}_{\text{tot}}(t). \tag{267}$$

The equation for the *operator* $b^\dagger$ may be considered as the quantum mechanical analogue of the van der Pol equation in the rotating wave approximation with a noisy driving force $(1/\gamma)\,\tilde{F}_{\text{tot}}(t)$. In the following Section 4.6 we solve the exact eqns (253) and (254) rather than the approximate eqn (267) by the method of quantum mechanical quasilinearization, because no substantial additional difficulties arise. On the other hand the final form of the Fokker–Planck equation near laser threshold can be obtained in a more or less heuristic way directly from (267), if this equation is interpreted as a classical (c-number) equation. We shall follow up this line in Section (4.12.c) so that the reader may proceed after having read Section 4.6 right away to Section (4.12.c), unless he is interested in the rigorous quantum mechanical treatment.

4.6. QUANTITATIVE TREATMENT OF SINGLE MODE OPERATION (QUANTUM MECHANICAL QUASI-LINEARIZATION)[26,54]

Somewhat above threshold the eqns (253) and (254) can be linearized by the ansatz (260), keeping only terms linear in ρ and ϕ. Note that in the quantum mechanical treatment ρ and ϕ are still operators while r_0 is a c-number (in these lectures we refrain from a representation of the precautions which this operator treatment requires). We first eliminate S in eqn (254) by inserting the expression (266) into eqn (253). Then we introduce the ansatz (260) and split the resulting equation for r_0, ρ and ϕ into its "real" and "imaginary" parts. In zeroth order, i.e. no fluctuations present, we obtain an equation for r_0, which is identical with that of the photon number derived in the rate equation treatment, see (21) with the spontaneous emission rate $W = g^2/\gamma$. The terms *linear* in the fluctuations give rise to the phase-fluctuations (imaginary part) and the amplitude fluctuations (real part).

If the laser mode is in resonance with the atomic transition frequency, the equations for ρ and ϕ are uncoupled, so that we may discuss these equations separately.

Phase noise, linewidth formula

The equation for ϕ reads

$$\ddot{\phi}+(\kappa+\gamma)\,\dot{\phi} = \frac{1}{r_0}\,\mathrm{Im}\{\exp(-i\phi)(\gamma\tilde{F}^{\dagger}+\dot{\tilde{F}}^{\dagger}+ig\,\tilde{\Gamma}_{21})\}. \tag{268}$$

The factor $\exp(-i\phi)$ represents a strong nonlinearity. It can be absorbed into the fluctuating forces because, as can be shown, the fluctuating forces can be represented by random pushes with random phases. Thus $\exp(i\phi)$ can be combined to a new random phase factor $\exp[i(\phi+\phi_j)$. Equation (268) is readily integrated. As we will show at the end of this paragraph the linewidth (half-width at half-power) due to phase noise is given by

$$\Delta\nu = \frac{1}{2t}\langle(\phi(t)-\phi(0))^2\rangle, \qquad [t > 1/(\gamma+\kappa)] \tag{269}$$

where the average refers to all coordinates of the noise sources. Using the properties of the noise sources F, Γ one obtains

$$\Delta\nu = \frac{1}{2r_0{}^2(\kappa+\gamma)^2}\{\tfrac{1}{2}g^2\,\gamma(N_1+N_2)+\kappa\gamma^2(n_{\mathrm{th}}+\tfrac{1}{2})\}, \tag{270}$$

where $N_2 = N\langle\rho_{22}\rangle$, $N_1 = N\langle\rho_{11}\rangle$ is the mean occupation number of levels 2 and 1, respectively, of all atoms and n_{th} is the thermal photon number. We eliminate the coupling constant g, which specializes in the present case of an exact resonance to the equation

$$g^2 = \kappa\gamma/(N_2-N_1), \tag{271}$$

(note that N_j is the actual occupation number under the joint action of "heat-baths" and laser field). Thus we find, finally, for the linewidth (halfwidth at half power)

$$\Delta\nu = \frac{\gamma^2\,\kappa^2}{(\kappa+\gamma)^2}\,\frac{\hbar\omega}{P}\left\{\frac{1}{2}\,\frac{N_2+N_1}{N_2-N_1}+n_{\mathrm{th}}+\frac{1}{2}\right\}, \tag{272}$$

where $P = 2\kappa r_0{}^2\hbar\omega$ is the output power, ω is the mode frequency, γ and κ are the atomic and the cavity halfwidth, respectively.

The term $(N_2+N_1)/2(N_2-N_1)$ stems from the atomic noise sources and expresses spontaneous emission noise. The term $n_{\mathrm{th}}+\frac{1}{2}$ arises from thermal resonator noise and vacuum fluctuations and stems from the field noise source $F(t)$. It was soon recognized by Shimoda *et al.* that both thermal noise quanta and spontaneous emission noise quanta should occur in a symmetric manner in $\Delta\nu$. This is now achieved by using the complete set of noise sources. In the maser, the thermal number of photons n_{th} is much bigger than unity (because

$kT \gg \hbar\omega$) so that the thermal noise of the resonator determines the linewidth. In the laser this contribution is entirely negligible and $(N_2+N_1)/(N_2-N_1)>1$ is dominant. In a two-level system it is many orders of magnitude bigger than the other two terms. The expressions for the linewidth above and below threshold differ by a factor $\frac{1}{2}$ in the quantum mechanical case as they do in the classical case.

The lineshape is a Lorentzian which is cut off at frequencies $\approx \kappa + \gamma$. In order to derive the lineshape as well as the relation (269) we consider the coherence function

$$\langle b^\dagger(t)\, b(0)\rangle, \tag{273}$$

where $\exp(i\omega t)$ is split off. We insert

$$b^\dagger \approx r_0 \exp[i\phi(t)] \tag{274}$$

into (273) and use the fact that both $\Gamma(= \sum \Gamma_\mu)$ and F consists of many statistically (or quantum mechanically) independent systems (i.e. they are Gaussian), so that

$$\phi(t)-\phi(0) = \sum_\mu \phi_\mu \tag{275}$$

$$\langle b^\dagger(t)\, b(0)\rangle \approx r_0{}^2 \prod_\mu \langle \exp[i\phi_\mu(t)]\rangle \approx r_0{}^2 \prod_\mu \{1+i\langle\phi_\mu(t)\rangle - \tfrac{1}{2}\langle\phi_\mu(t)^2\rangle\}. \tag{276}$$

where ϕ is the fluctuating phase of the mode and ϕ_0 is the initial value of the phase. Because

$$\langle\phi_\mu\rangle = 0 \tag{277}$$

and thus

$$\sum_\mu \langle\phi_\mu{}^2\rangle = \langle(\phi-\phi_0)^2\rangle, \tag{278}$$

we have finally

$$\langle b^\dagger(t)\, b(0)\rangle = r_0{}^2 \exp\{-\tfrac{1}{2}\langle(\phi-\phi_0)^2\rangle\}. \tag{279}$$

The explicit evaluation of (278), which we performed above, has shown that the quantity $\langle(\phi-\phi_0)^2\rangle$ is proportional to the time t. On the other hand we should expect that for a classical electric field with a Lorentzian line-shape we have $E(t) \propto \exp(-\Delta\nu t)$ where $\Delta\nu$ is the half-width at half power. Thus the laser line-shape is indeed a Lorentzian. The above line-width formula holds in conditions well above threshold, i.e.

$$\langle\rho^2\rangle \ll r_0{}^2,$$

where ρ is an amplitude fluctuation.

Amplitude fluctuations

We start again with the linearized equation where we compare now the "real" parts of both sides (again after multiplication with $\exp[-i\phi(t)]$). This yields the following equation

$$\ddot{\rho}+(\kappa+\gamma)\dot{\rho}-2|g|^2 r_0{}^2\int^t(\dot{\rho}+2\kappa\rho)K_1(t,\tau)\,\mathrm{d}\tau$$

$$=-|g|^2 r_0\left[\int^t\left(r_0(\tilde{F}+\tilde{F}^\dagger)+\tfrac{1}{2}(\Gamma_{22}-\Gamma_{11})\right)K_1(t,\tau)\right]$$

$$+\mathrm{Re}\{\exp(-i\phi)(\gamma\tilde{F}^\dagger+\dot{\tilde{F}}^\dagger+ig^*\tilde{\Gamma}_{21}\},\qquad(280)$$

where $K_1(t,\tau)=-2\exp[-(t-\tau)/T]$ is the integral kernel which stems from the elimination of S in (253) and (254). For the solution we perform a Fourier-transformation

$$\rho(t)=\int_{-\infty}^{+\infty}\exp(i\omega t)\,\rho(\omega)\,\mathrm{d}\omega.\qquad(281)$$

We then obtain readily for the Fourier-transform of eqn (280)

$$\rho(\omega)N(\omega)=M(\omega),\qquad(282)$$

where

$$N(\omega)=\{-\omega^2+i\omega(\kappa+\gamma)-2g^2 r_0(i\omega+2\kappa)K_1(\omega)\}\qquad(283)$$

with

$$K_1(\omega)=-\frac{2}{w_{12}+w_{21}+i\omega}.\qquad(284)$$

The polynomial $M(\omega)$ is of a complicated structure, so that we drop this intermediate result. If $N(\omega)$ possesses one or several real roots ω_j, then the modulation of ρ of $|b|=r_0+\rho$ shows an undamped oscillation ("spiking"). A closer discussion of $N(\omega)$ shows that a necessary condition for undamped spiking is

$$\gamma+\tfrac{1}{2}w_{21}+w_{12}<\kappa,\qquad(285)$$

which holds only in exceptional cases however. For the experimentally observed undamped spiking, other mechanisms must therefore also be invoked. Because the calculation of the correlation functions of the amplitude fluctuations is elementary, but lengthy, we only quote the final result

$$\langle\rho(t)\,\rho(t+\tau)\rangle=\frac{1}{2\pi}\int_{-\infty}^{+\infty}\mathrm{d}\omega\,\frac{G(\omega)\exp(i\omega\tau)}{h(\omega)\,h(-\omega)},\qquad(286)$$

where $G(\omega)$ is a polynomial of ω of fourth order, while the h's are polynomials of third order. For moderate photon numbers, i.e.

$$r_0 g < w_{ij}, \gamma, \tag{287}$$

the expression for the intensity fluctuations is found to be

$$k(\tau) \equiv \langle b^\dagger(t)\, b^\dagger(t+\tau)\, b(t+\tau)\, b(t)\rangle - \langle b^\dagger b\rangle_t \langle b^\dagger b\rangle_{t+\tau}$$

$$= \frac{1}{2}\left(\frac{1}{d_{\text{thr}}} + 2n_{\text{th}} + 1\right) \frac{d_{\text{thr}}\, {r_0}^2}{d_0 - d_{\text{thr}}} \exp[-2\kappa\tau(d_0 - d_{\text{thr}})/d_{\text{thr}}], \tag{288}$$

where d_{thr} is the (atomic) inversion at threshold, d_0 is the unsaturated inversion which one would obtain for a given pump rate without laser action. Formula (288) holds also for inhomogeneously broadened solid state and gas lasers[31] in a region above threshold which starts with a photon number ${r_0}^2$ which equals 5–10 times the photon number at threshold up to photon numbers satisfying (287). Because $d_0 - d_{\text{thr}}$ is proportional to the photon number ${r_0}^2$, the frequency of the intensity fluctuations increases with ${r_0}^2$, whereas (286) decreases according to $1/{r_0}^2$. Both predictions are fully substantiated experimentally in a region ${r_0}^2 \geqslant 10 \times ({r_0}^2)_{\text{thr}}$.

4.7. THE DENSITY MATRIX EQUATION FOR A SET OF 2-LEVEL ATOMS AND A FIELD MODE[55]

In this chapter we sketch the method by which to derive the density matrix equation and write it down explicitly for 2-level atoms and a single mode. The reader, who is interested in the general derivation of the density matrix equation for an arbitrary system is referred to in Section 5.

We consider the interaction of the light mode or the electron of an atom with a heatbath or a set of heatbaths. The density matrix W of the total system obeys the equation†

$$\frac{\mathrm{d}W}{\mathrm{d}t} = -\frac{i}{\hbar}[H, W], \tag{289}$$

where

$$H = H_0 + H_{FB} \tag{290}$$

and

$$H_0 = H_F + H_B, \tag{291}$$

† In this section we abbreviate H (field-bath) and H (electron-bath) by H_{FB}, $H_{(\text{field})}$ or $H_{(\text{electron})}$ by H_F and H(bath) by H_B.

The Hamiltonian of the free field is H_F and is

$$H_F = \begin{cases} \hbar\omega b^\dagger b & \text{light mode} \quad (292) \\ \hbar\nu a_2{}^\dagger a_2 & \text{atom} \quad (293) \end{cases}$$

The Hamiltonian of the heatbath H_B may consist of a sum of several Hamiltonians corresponding to different heatbaths

$$H_B = \sum_j H_B{}^{(j)}. \quad (294)$$

H_{FB} represents the interaction between the two systems. For our analysis it is convenient to proceed to the interaction representation

$$\tilde{W} = \exp(iH_0 t/\hbar)\, W \exp[(-iH_0 t/\hbar)] \quad (295)$$

$$\tilde{H}_{FB} = \exp(iH_0 t/\hbar)\, H_{FB} \exp(-iH_0 t/\hbar). \quad (296)$$

The interaction Hamiltonian has the form

$$H_{FB} = \hbar \sum V_k(t)\, B_k(t) = \begin{cases} \underbrace{b\, e^{-i\omega t}}_{V_1}\ \underbrace{B^\dagger(t)}_{B_1} + \underbrace{b^\dagger e^{i\omega t}}_{V_2}\ \underbrace{B(t)}_{B_2} & \text{light mode} \quad (297)\dagger \\ \underbrace{a_2{}^\dagger a_1\, e^{i\nu t}}_{V_1}\ \underbrace{B_{21}(t)}_{B_1} + \underbrace{a_1{}^\dagger a_2\, e^{-i\nu t}}_{V_2}\ \underbrace{B_{12}(t)}_{B_2} & \text{electron} \quad (298) \end{cases}$$

where the V's are field or electron operator's and the B's are bath operators. The explicit form of the V's suggests that we write

$$V_k(t) = V_k(0) \exp(i\Delta\nu_k t).$$

The idea for the derivation of the density matrix is now this. We solve eqn (289) in the interaction representation by perturbation theory up to second order

$$\tilde{W} = W_0 + \left(-\frac{i}{\hbar}\right)\int_0^t d\tau\, [\tilde{H}_{FB}, W_0] + \left(-\frac{i}{\hbar}\right)^2 \int_0^t d\tau_2 \int_0^{\tau_2} d\tau_1 \{\ldots\} \quad (299)$$

$$\{\ldots\} = [\tilde{H}_{FB}(\tau_2), [\tilde{H}_{FB}(\tau_1), W_0]].$$

W_0 is the density matrix of the total system at time $t = 0$.

We then take, on both sides, the average over the heatbaths which are supposed to be kept at certain given temperatures. Assuming that the average

† Here and in the following we include only heatbaths causing *real* transitions.

over the heatbath variables vanishes, i.e. $\bar{B} = 0$, we obtain the following result

$$\tilde{\rho} = \tilde{\rho}_0 + \left(-\frac{i}{\hbar}\right)^2 \int_0^t \mathrm{d}\tau_2 \int_0^{t_2} \mathrm{d}\tau_1 \overline{\{\ldots\}}, \tag{300}$$

where $\begin{cases} \overline{\{\ldots\}} = \sum_{kk'} \overline{[V_k(\tau_2)\, B_k(\tau_2), [V_{k'}(\tau_1)\, B_{k'}(\tau_1), W_0]]}. \\ \bar{\bar{W}} = \tilde{\rho} \end{cases}$

and $\tilde{\rho}$, $\tilde{\rho}_0$ are the density matrices of the system averaged over reservoir variables.

We now assume that the heatbaths have a short memory. This implies that

$$\overline{B(\tau_2)\, B(\tau_1)} \sim \delta(\tau_2 - \tau_1), \tag{301}$$

so that the double integral reduces to a single integral which may be approximated by the product of the integrand and the time t if t is small enough. It must be noted that the remaining operators V_k still act on the density matrix $\tilde{\rho}_0$.

A closer inspection shows that the resulting right-hand side can be written in the form of commutators (see below). We now bring $\tilde{\rho}_0$ to the left-hand side and divide both sides by t. Taking the limit of small t we may put

$$(\tilde{\rho} - \tilde{\rho}_0)/t = \mathrm{d}\tilde{\rho}/\mathrm{d}t. \tag{302}$$

Assuming further that we consecutively repeat the step just done, we may insert an arbitrary time into $\tilde{\rho}_0$. This leaves us with the equation

$$\frac{\mathrm{d}\tilde{\rho}(t)}{\mathrm{d}t} = \sum_{kk'} \{[V_{k'}\, \tilde{\rho}(t), V_k]\, A_{kk'} + [V_{k'}, \rho(t)\, V_k]\, A'_{kk'}\}. \tag{303}$$

The coefficients A, A' contain the correlation functions of the heatbath variables. A detailed discussion shows that only those terms k, k' contribute for which $\Delta\nu_k = -\Delta\nu_{k'}$, so that we use only those terms further on.

Specialization of eqn (303)[56]

a. Light mode. We identify $V_1 = b \exp(-i\omega t)$ and $V_2 = b^\dagger \exp(i\omega t)$. Using new abbreviations for the coefficients A, the density matrix equation reads

$$\frac{\mathrm{d}\rho}{\mathrm{d}t} = \delta([b^\dagger \rho, b] + [b^\dagger, \rho b]) + \xi([b\rho, b^\dagger] + [b, \rho b^\dagger]). \tag{304}$$

We first show how this equation allows us to treat the mean motion of the light field amplitude and of the intensity, where we define the average of an arbitrary operator O by

$$\langle O \rangle = \mathrm{tr}(O\rho). \tag{305}$$

The average lightfield amplitude obeys the equation

$$\frac{\mathrm{d}\langle b^\dagger\rangle}{\mathrm{d}t} = \{i\omega-(\xi-\delta)\}\langle b^\dagger\rangle. \tag{306}$$

where δ and ξ are constants.

The photon number obeys the equation

$$\frac{\mathrm{d}\langle b^\dagger b\rangle}{\mathrm{d}t} = 2\delta-2(\xi-\delta)\langle b^\dagger b\rangle. \tag{307}$$

Integrating (306), we get

$$\langle b^\dagger\rangle = \langle b^\dagger\rangle_0 \exp[i\omega t-(\xi-\delta)t]. \tag{308}$$

The solution of (307) is

$$\langle b^\dagger b\rangle = \langle b^\dagger b\rangle_0 \exp[-2(\xi-\delta)t]+\frac{\delta}{\xi-\delta}(1-\exp[-2(\xi-\delta)t]). \tag{309}$$

Evidently the difference $\xi-\delta$ must be identified with the decay constant which is introduced in the classical theory

$$\xi-\delta = \kappa, \tag{310}$$

whereas the expressions $\delta/(\xi-\delta)$ must be identified with the photon number $n(T)$ at the equilibrium temperature, T.

b. Atom. We make the following identifications in (298)

$$V_1 = a_2^\dagger a_1 \exp(ivt), \qquad V_2 = a_1^\dagger a_2 \exp(-ivt).$$

The density matrix equation is

$$\frac{\mathrm{d}\tilde{\rho}}{\mathrm{d}t} = \frac{w_{21}}{2}\{[a_1^\dagger a_2, \tilde{\rho}a_2^\dagger a_1]+[a_1^\dagger a_2\,\tilde{\rho}, a_2^\dagger a_1]\}$$

$$+\frac{w_{12}}{2}\{[a_2^\dagger a_1, \tilde{\rho}a_1^\dagger a_2]+[a_2^\dagger a_1\,\tilde{\rho}, a_1^\dagger a_2]\}. \tag{311}$$

Using the notation w_{21}, w_{12} instead of $A_{kk'}$, $A'_{kk'}$, we have anticipated that these quantities are just the transition rates used earlier. To demonstrate this we investigate the rate of change of the average of occupation numbers,

$$\langle a_2^\dagger a_2\rangle = \mathrm{tr}(a_2^\dagger a_2\rho) \tag{312}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle a_2^\dagger a_2\rangle = \left\langle a_2^\dagger a_2 \frac{\mathrm{d}\tilde{\rho}}{\mathrm{d}t}\right\rangle. \tag{313}$$

Inserting for $\mathrm{d}\tilde{\rho}/\mathrm{d}t$ on the right-hand side, using the cyclic property of traces,

and using the relation $a_i^\dagger a_k \,.\, a_l^\dagger a_m = \delta_{kl} a_i^\dagger a_m$ (for one electron) we readily obtain

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle a_2^\dagger a_2\rangle = w_{12}\langle a_1^\dagger a_1\rangle - w_{21}\langle a_2^\dagger a_2\rangle. \tag{314}$$

In the same way we find

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle a_1^\dagger a_2\rangle = -\tfrac{1}{2}(w_{12}+w_{21})\langle a_1^\dagger a_2\rangle, \tag{315}$$

so that in the present model we have

$$2\gamma = w_{12}+w_{21}. \tag{316}$$

c. Field mode coupled to many atoms. It is a simple matter to extend the above results, a and b, to the joint system of a field mode and many atoms. One may show that the expressions for $(\mathrm{d}\tilde{\rho}/\mathrm{d}t)_{\text{field}}$ and $(\mathrm{d}\tilde{\rho}/\mathrm{d}t)_{\text{atom}}$ just add to each other. Returning from the interaction representation to the Schrödinger picture and introducing the coherent interaction Hamiltonian (204) we find

$$\frac{\mathrm{d}\rho}{\mathrm{d}t} = -\frac{i}{\hbar}[H_F, \rho] + \left(\frac{\partial\rho}{\partial t}\right)_{\text{field}} + \sum_{\mu}\left(\frac{\partial\rho}{\partial t}\right)_{\text{atom }\mu}, \tag{317}$$

where H_F and $(\partial\rho/\partial t)_{\text{field}}$ are given by (206) and (304) respectively. The right-hand side of eqn (311) gives $(\partial\rho/\partial t)_{\text{atom}}$, provided the operators $a_i^\dagger a_j$ are supplemented by the atomic index μ.

4.8. THE EVALUATION OF MULTI-TIME CORRELATION FUNCTIONS BY THE SINGLE TIME DENSITY MATRIX[57]

We shall show that the density matrix which contains only one time variable may be used to calculate multi-time correlation functions. It is known (see Glauber's chapter in this volume) that the statistical properties (or, in other words, the coherence properties) of laser light are determined by correlation functions of the form

$$\langle b^\dagger(t_1')\ldots b^\dagger(t_m')\, b(t_n)\ldots b(t_1)\rangle, \tag{318}$$

where we consider a single mode for simplicity. However, all our following considerations apply equally well to the multimode case. We assume, that the b's occur ordered with respect to their time

$$t_{j+1} \geqslant t_j; \qquad t'_{j+1} \geqslant t'_j. \tag{319}$$

In order to show how (318) can be calculated by the density matrix, we represent (318) in the form

$$\mathrm{tr}\{b(t_n)\, b(t_{n-1}) \ldots b(t_1)\, \rho(t_0)\, b^\dagger(t_1') \ldots b^\dagger(t_m')\}, \tag{320}$$

where $\rho(t_0)$ is the density matrix at time $t_0 = \min(t_j, t_j')$. We can rewrite expression (320) in a new fashion by means of functional derivatives, δ,

$$\frac{1}{(-i)^n}\,\frac{1}{i^m}\left[\frac{\delta^n}{\delta\alpha(t_n)\,\delta\alpha(t_{n-1})\ldots\delta\alpha(t_1)}\cdot\frac{\delta^m}{\delta\gamma^*(t_1')\ldots\delta\gamma^*(t_m')}\,\rho(\alpha,\gamma)\right]_{\substack{\alpha\equiv 0\\ \gamma\equiv 0}} \tag{321}$$

where

$$\begin{aligned}\rho(\alpha,\gamma) = {} & \left\langle T\exp\left\{-i\int_{t_0}^{t} H_\tau\,\mathrm{d}\tau - i\int_{t_0}^{t}\left(\alpha(\tau)\,b_\tau + \alpha^*(\tau)\,b_\tau^\dagger\right)\mathrm{d}\tau\right\}\right. \\ & \left.\times\,\rho(t_0)\,\tilde{T}\exp\left\{i\int_{t_0}^{t} H_{\tau'}\,\mathrm{d}\tau' + i\int_{t_0}^{t}\left(\gamma(\tau')\,b_{\tau'} + \gamma^*(\tau')\,b_{\tau'}^\dagger\right)\mathrm{d}\tau'\right\}\right\rangle_{\text{reservoir}}\end{aligned} \tag{322}$$

Here H_τ, and $H_{\tau'}$ are Hamiltonians at times τ, τ', and α, γ are arbitrary functions. The time ordering operators, T and $\tilde{T}$ order the terms with latest time to the left and to the right, respectively.

The total Hamiltonian, H, of the system comprises the light field and its interaction with the atoms, the interaction between the light field and heatbaths, and the interaction between the atoms and the heatbaths, thus we have

$$\begin{aligned} & H = H_S + H_{S-B}, \\ \text{where} \quad & H_S = H_{\text{field}} + H_{\text{atoms}} + H_{\text{field-atom}} \\ \text{and} \quad & H_{S-B} = H_{\text{field-bath}} + H_{\text{atoms-bath}}. \end{aligned} \tag{323}$$

It is assumed that the free motion of the heatbaths is split off, so that H_τ contains the coordinates of the heatbaths in the interaction representation. The problem is now reduced to the determination of the *extended* density matrix (322). By differentiation of both sides of (322) with respect to time we obtain an equation for $\rho(\alpha, \gamma)$.

$$\begin{aligned}\frac{\mathrm{d}\rho(\alpha,\gamma)}{\mathrm{d}t} = {} & -i[H_S, \rho] - i\left(\alpha(t)\,b + \alpha^*(t)\,b^\dagger\right)\rho + \rho i\left(\gamma(t)\,b + \gamma^*(t)\,b^\dagger\right) \\ & - i\langle[H_{SB}, \tilde{\rho}]\rangle_{\text{res}}.\end{aligned} \tag{324}$$

The right-hand side of (322) defines $\tilde{\rho}$ but without the reservoir average.

The last term calculated from the above, coincides with

$$\left(\frac{\partial\rho}{\partial t}\right)_{\text{incoh. light}} + \left(\frac{\partial\rho}{\partial t}\right)_{\text{incoh. atom}}$$

on the right-hand side of eqn (317). Therefore, the only difference between the usual density matrix and the new equation (324) consists in the additional terms containing the α's and γ's which are arbitrary functions (which may be small). In order to determine the correlation function (318), the system must be "probed" by classical external fields.† The b's in (324) are, of course, time independent. If we want to calculate only a limited number of correlation functions, it is sufficient to formulate an initial value problem. For this we put

$$\alpha(t) = \sum_i \alpha_i \, \delta(t - t_i), \qquad \gamma^*(t) = \sum_i \gamma_i^* \, \delta(t - t_i') \tag{325}$$

and replace the functional derivative by an ordinary derivative with respect to α. Here t_i is the time at which the jump occurs. Obviously the solution of (324) is now replaced by solving the usual density matrix equation (317) with the additional jump conditions given by

$$\begin{aligned} \left.\frac{\mathrm{d}\rho}{\mathrm{d}t}\right|_{t=t_i} &= \rho(t_i+0) - \rho(t_i-0) = -i\alpha b \int_{t_i-\varepsilon}^{t_i+\varepsilon} \delta(t'-t_i)\, \rho(t')\, \mathrm{d}t' \\ &= -i\alpha b \rho(t_i - 0) + \text{correction terms of order } \alpha^2 \end{aligned} \tag{326}$$

The corrections of order α^2 and higher order may be neglected as long as the differentiation in eqn (326) is performed only once with respect to time at a given time. The case that several operators occur in (318) at the same time may be then obtained as a special case by letting the time difference go to zero.

So far we have been considering correlation functions with respect to photon operators. In a similar way, we may do this, of course, with atomic operators or with mixed terms, for instance, with

$$\langle (a_{i_1'}^\dagger \, a_{k_1'})(t_1') \ldots (a_{i_{m'}}^\dagger \, a_{k_{m'}})(t_m')(a_{i_n}^\dagger \, a_{k_n})(t_n) \ldots (a_{i_1}^\dagger \, a_{k_1})(t_1) \rangle, \tag{327}$$

where the $a_i^\dagger$ and a_j are the creation and annihilation operators of electrons. Obviously one has now a density matrix equation of the form

$$\begin{aligned} \frac{\mathrm{d}\rho}{\mathrm{d}t} &= -i[H_S, \rho] + \Lambda_1 \rho + \rho \Lambda_2 - i \sum_{ik} D_{ik}(t)\, a_i^\dagger \, a_k \, \rho \\ &\quad + \rho i \sum_{ik} D'_{ik}(t)\, a_i^\dagger \, a_k - i\big(\alpha(t)\, b + \alpha^*(t)\, b^\dagger\big)\, \rho + i\rho\big(\gamma(t)\, b + \gamma^*(t)\, b^\dagger\big) \end{aligned} \tag{328}$$

and the correlation function (327) may be found by functional derivatives with respect to the coefficients D.

† This establishes a connection with Kubo's point of view.[58]

4.9. GENERALIZED FOKKER–PLANCK EQUATION; DEFINITION OF DISTRIBUTION FUNCTIONS

a. For the field

(*i*) *Wigner distribution function and related representations.* The Wigner distribution function [59] establishes a connection between the quantum mechanical wave function, or more generally, the density matrix, and a *c*-number distribution function of (classical) variables ($u\beta$). The Wigner function is

$$f(u_1, u_2) = \int\int \exp(-iu_1\beta_1 - iu_2\beta_2)\,\mathrm{tr}\{\exp(i\beta_1 x + i\beta_2 p)\rho\}\,\mathrm{d}\beta_1\,\mathrm{d}\beta_2, \quad (329)$$

where x and p are coordinate and momentum operators,† respectively, of a harmonic oscillator, and ρ is the density matrix.

Equation (329) establishes a connection between the quantum mechanical variables x and p and classical variables u_1, u_2:

classical	transformation (329)	quantum mechanical
u_1	$\longleftrightarrow$	x
u_2	$\longleftrightarrow$	$p = \frac{\hbar}{i}\frac{\mathrm{d}}{\mathrm{d}x}$

(330)

Equation (329) corresponds to the definition of the distribution function in probability theory of classical variables where x and p would be random functions and $\mathrm{tr}[\exp(i\beta_1 x + i\beta_2 p)\rho]$ would be just the characteristic function. The definition is not confined to an harmonic oscillator, but can also be used for other quantum systems. In the following we leave it open whether ρ or (x, p) are time dependent operators, because (329) holds for both the Schrödinger and Heisenberg representation.

In our present context it is advantageous to proceed from (x, p) of the harmonic oscillator to the corresponding Bose creation and annihilation operators $b^\dagger, b$ by the equations

$$x = (b^\dagger + b)\sqrt{\frac{\hbar}{2\omega}}$$

$$p = i(b^\dagger - b)\sqrt{\frac{\hbar\omega}{2}}. \quad (331)$$

Inserting (331) into (329) yields

$$\int \exp(-i\beta u - i\beta^* u^*)\,\mathrm{tr}\{\exp(i\beta b + i\beta^* b^\dagger)\rho\}\,\mathrm{d}^2\beta, \quad (332)$$

† x and p are defined in such a way that the Hamilton operator becomes $H = \frac{1}{2}(p^2 + \omega^2 q^2)$.

where we have introduced

$$\beta = \sqrt{\frac{\hbar}{2}}\left(\frac{\beta_1}{\sqrt{\omega}} - i\omega^{\frac{1}{2}}\beta_2\right) \qquad u = \frac{\omega u_1 + iu_2}{\sqrt{(2\omega\hbar)}} \tag{333}$$

$$\mathrm{d}^2\beta = \mathrm{d}(\mathrm{Re}\,\beta)\,\mathrm{d}(\mathrm{Im}\,\beta)$$

Using Feynman's disentangling techniques[60] we obtain

$$\exp(i\beta b + i\beta^* b^\dagger) = \exp(|\beta|^2/2)\exp(i\beta b)\exp(i\beta^* b^\dagger) \tag{334}$$

The sequence of the operators $\exp(i\beta b)$ and $\exp(i\beta^* b^\dagger)$ is rather unimportant because we have

$$\exp(i\beta^* b^\dagger)\exp(i\beta b) = \exp(i\beta b)\exp(i\beta^* b^\dagger)\exp|\beta|^2 \tag{335}$$

There are thus two equivalent forms of (332):

$$W(u,u^*) = \frac{1}{\pi^2}\int \exp(-i\beta u - i\beta^* u^*)\,\mathrm{tr}\{\exp(i\beta b)\exp(i\beta^* b^\dagger)\rho\}\exp(|\beta|^2/2)\mathrm{d}^2\beta \tag{336}$$

$$= \frac{1}{\pi^2}\int \exp(-i\beta u - i\beta^* u^*)\,\mathrm{tr}\{\exp(i\beta^* b^\dagger)\exp(i\beta b)\rho\}\exp(-|\beta|^2/2)\mathrm{d}^2\beta \tag{337}$$

where the Wigner distribution function, $W(u,u^*)$, is now normalized with respect to the complex u-plane.

Somewhat different definitions† for distribution functions are obtained by omitting the factors $\exp(\pm|\beta^2|/2)$

$$P(u,u^*) = \frac{1}{\pi^2}\int\int \exp(-i\beta u - i\beta^* u^*)\mathrm{tr}\{\exp(i\beta^* b^\dagger)\exp(i\beta b)\,\rho\}\mathrm{d}^2\beta \tag{338a}$$

$$Q(u,u^*) = \frac{1}{\pi^2}\int\int \exp(-i\beta u - i\beta^* u^*)\,\mathrm{tr}\{\exp(i\beta b)\exp(i\beta^* b^\dagger)\,\rho\}\mathrm{d}^2\beta. \tag{338b}$$

Introducing coherent states $|u\rangle$ by the equation

$$b|u\rangle = u|u\rangle, \tag{339}$$

we can prove that $P(u,u^*)$ is the Glauber and Sudarshan[61] diagonal representation of the density operator ρ, given by

$$\rho = \int P(u,u^*)\,|u><u|\,\mathrm{d}^2u, \tag{340}$$

† All definitions of W, P, Q in this section secure that W, P, Q are real provided P is a hermitian operator. Note, that P, W are not necessarily positive, however. Therefore P, W do not have the usual meaning of a probability distribution, but they allow us to calculate all quantum mechanical expectation values purely by c-number procedures (see below).

whereas $Q(u, u^*)$ is the diagonal matrix element of the density operator in coherent states $|u\rangle$:

$$Q(u, u^*) = \frac{1}{\pi} \langle u|\rho|u\rangle. \tag{341}$$

For proof of (340) we insert this representation into the right-hand side of (338a) and use the cyclic property of the trace $\mathrm{tr}(AB) = \mathrm{tr}(BA)$. We then get

$$\frac{1}{\pi^2} \int\int\int\int P(u', u'^*) \exp[-i\beta(u-u') - i\beta^*(u^*-u'^*)]\, \mathrm{d}^2u'\, \mathrm{d}^2\beta$$

which is equal to $P(u, u^*)$. To prove (341) we insert the coherent state representation of the unity operator

$$1 = \frac{1}{\pi} \int |u\rangle \langle u|\, \mathrm{d}^2u$$

within the trace, leading to

$$\mathrm{tr}\left\{\exp(i\beta b) \exp(i\beta^* b^\dagger) \frac{1}{\pi} \int |u\rangle \langle u|\, \mathrm{d}^2u\, \rho\right\} = \frac{1}{\pi} \int \exp(i\beta u + i\beta^* u^*) \langle u|\rho|u\rangle \mathrm{d}^2u$$

Inserting this expression at the right-hand side of (339) and making use of the δ-function representation we immediately get (341). Due to the close relationship of the functions W, P, Q there exist equations connecting W, P, Q that read, according to Glauber,

$$Q(u, u^*) = \frac{2}{\pi} \int \exp(-2|u-\alpha|^2)\, W(\alpha, \alpha^*)\, \mathrm{d}^2\alpha$$

$$W(u, u^*) = \frac{2}{\pi} \int \exp(-2|u-\alpha|^2)\, P(\alpha, \alpha^*)\, \mathrm{d}^2\alpha$$

$$Q(u, u^*) = \frac{1}{\pi} \int \exp(-|u-\alpha|^2)\, P(\alpha, \alpha^*)\, \mathrm{d}^2\alpha.$$

(*ii*) *Transforms of the Wigner distribution function: characteristic functions.* Depending on the problem, one may use the Fourier-transform of the Wigner-function (329) instead of (329). This new function may be called the characteristic function (in analogy to classical statistics). It is given by

$$\chi_W(\beta_1, \beta_2) = \mathrm{tr}\{\exp(i\beta_1 x + i\beta_2 p)\rho\} \tag{342}$$

(apart from a constant factor[62]). Similarly, the characteristic functions belonging to (338) are

$$\chi_P(\beta, \beta^*) = \mathrm{tr}\{\exp(i\beta^* b^\dagger)\exp(i\beta b)\rho\} \tag{343a}$$

$$\chi_Q(\beta, \beta^*) = \mathrm{tr}\{\exp(i\beta b)\exp(i\beta^* b^\dagger)\rho\} \tag{343b}$$

Note, that these latter functions are Fourier transforms of (338) but not of (332), because of the factor $\exp(\pm|\beta|^2/2)$ (see (337)).

b. For electrons

(*i*) *Distribution functions for a single electron.* Schmid and Risken[63] have defined the distribution function

$$f(v_{ik}) = N' \int \ldots \int \exp\left[-i\sum_{jk} v_{jk}\,\xi_{jk}\right] \mathrm{tr}\left\{\exp\left[i\sum_{jk}\xi_{jk}\,a_j^\dagger a_k\right]\rho\right\} \mathrm{d}\{\xi\} \tag{344}$$

with

$$v_{ik} = v_{ki}^*.$$

Here the v's are classical variables associated with electron operators and N' is a normalization constant, so that

$$\int \ldots \int f(v)\,\mathrm{d}v = 1.$$

According to Haken *et al.*[64] another possibility is given by

$$f(v_{ik}) = N' \int \ldots \int \exp\left(-i\sum_{jk} v_{jk}\,\xi_{jk}\right) \mathrm{tr}\left(\prod_{j<k} \exp(i\,\xi_{jk}\,a_j^\dagger a_k)\right) \times \prod_k \exp(i\xi_k\,a_k^\dagger a_k) \prod_{j>k} \exp(i\xi_{jk}\,a_j^\dagger\,a_k)\,\mathrm{d}\{\xi\} \tag{345}$$

which has great advantages in practical calculations (see Section 4.10). When an equation of the Fokker–Planck type for the f's is derived in the case of a single atom it turns out that the f's are singular and that diffusion coefficients of high order are needed. In the actual laser one has to deal, however, with a great number of equivalent atoms. It is then possible to introduce macroscopic variables which show only small fluctuations, so that only the first terms of the generalized Fokker–Planck equation are important (see below). For a system of two-level atoms these macroscopic variables are defined as follows:

Macroscopic classical variables quantum mechanical operators

total dipole moment

$$v^* \longleftrightarrow S^+ = \sum_\mu (a_2^\dagger a_1)_\mu$$

$$v \longleftrightarrow S^- = \sum_\mu (a_1^\dagger a_2)_\mu \tag{346}$$

total inversion

$$D \longleftrightarrow 2S_z = \sum_\mu (a_2^\dagger a_2 - a_1^\dagger a_1)_\mu.$$

The above correspondence is established by the distribution function

$$f(v, v^*, D) = N' \int \ldots \int \exp(-i\{v\xi + v^*\xi^* + \zeta D/2\})\, \chi(\xi, \xi^*, \zeta)\, \mathrm{d}^2\xi\, \mathrm{d}\zeta. \tag{347}$$

where

$$\chi = \mathrm{tr}\{\exp(i\xi^* S^+) \exp(i\zeta S_z) \exp(i\xi S^-)\, \rho\}. \tag{348}$$

A trivial generalization consists in replacing ζS_z by $\xi_1 N_1 + \xi_2 N_2$ where N_1, N_2 are the total occupation numbers of levels 1 and 2.

(*ii*) *Characteristic functions.* These functions are defined by

$$\chi(\xi_{jk}) = \mathrm{tr}\left\{\exp\left(i \sum_{jk} \xi_{jk}\, a_j^\dagger a_k\right) \rho\right\}$$

or (349)

$$\chi(\xi_{jk}) = \mathrm{tr}\left\{\prod_{j<k} \exp(i\xi_{jk}\, a_j^\dagger a_k) \prod_k \exp(i\xi_k\, a_k^\dagger a_k) \prod_{j>k} \exp(i\xi_{jk}\, a_j^\dagger\, a_k)\, \rho\right\}$$

(*iii*) *Electrons and fields.* In this case it is possible to take any combination of the definition of (i) and (ii). In the following we shall consider the example

$$f(u, u^*, v, v^*, D) = N' \int \ldots \int \exp\left[-i\left(v\xi + v^*\,\xi^* + \zeta\frac{D}{2} + u\beta + u^*\beta^*\right)\right] \times \chi(\xi, \xi^*, \zeta, \beta, \beta^*)\, \mathrm{d}^2\xi\, \mathrm{d}\zeta\, \mathrm{d}^2\beta, \tag{350}$$

where

$$\chi = \mathrm{tr}\{O(\xi, \xi^*, \zeta, \beta, \beta^*)\, \rho(t)\}$$

and

$$O = \exp(i\xi^*\, S^+) \exp(i\zeta\, S_z) \exp(i\xi S^-) \exp(i\beta^*\, b^\dagger) \exp(i\beta b). \tag{351}$$

The characteristic function is obviously χ.

To find the equation of motion for the distribution function of the *field* it makes little difference whether we use the Heisenberg or the Schrödinger representation. In the Heisenberg representation the operators are time

dependent, whereas the density matrix is considered as time independent. On the other hand, in the Schrödinger representation, the operators are time independent, whereas the density matrix is time dependent. For the *atoms* it turns out, however, that the Heisenberg picture leads to very complicated expressions. So far, nobody has been able to derive a completely exact equation for the distribution function in that case (see e.g. Lax and Yuen[43]). On the other hand, the treatment of Haken *et al.*,[64] who used the Schrödinger representation and a product of exponential operators, has led to a completely exact equation for two-level atoms. We first give a sketch of how to derive this exact equation and then we present the results for the arbitrary dissipative quantum system which comprises as special cases an N-level atom (see Section 5).

4.10 GENERALIZED FOKKER–PLANCK EQUATION

Exact equation for the distribution function for a set of two level atoms[64]†

We differentiate both sides of (350) with respect to the time. Because χ depends on the time only, via ρ (see (351), we have

$$\frac{\mathrm{d}\chi}{\mathrm{d}t} = \mathrm{tr}\left\{O\frac{\mathrm{d}\rho}{\mathrm{d}t}\right\}. \tag{352}$$

When we introduce for $\mathrm{d}\rho/\mathrm{d}t$, the right-hand side of the density matrix, we obtain expressions of the form

$$\mathrm{tr}(Oa_2^{\dagger}\, a_2\, \rho), \quad \mathrm{tr}(Oa_2^{\dagger}\, a_1\, \rho a_1^{\dagger}\, a_2), \quad \mathrm{tr}(Ob\rho b^{\dagger}), \text{ etc.} \tag{353}$$

If $a_i^{\dagger}a_k$ and b were not operators, we could obtain, for example, the expression

$$\mathrm{tr}(O(a_2^{\dagger}\, a_2 - a_1^{\dagger}\, a_1)\rho), \tag{354}$$

by differentiating O with respect to ξ which occurs as a factor of $(a_2^{\dagger}\, a_2 - a_1^{\dagger}\, a_1)$ in the exponential. Because the $a_1^{\dagger}\, a_2$ are operators, the procedure is not quite so simple, because by commuting a pair of a's with another pair of a's usually new pairs are generated. However, these pairs form an algebraic ring which is finite. Therefore, it is, in fact, possible to replace all suitable expressions of the form (353) by derivatives of O with respect to classical c-numbers. Therefore all quantum mechanical procedures can be replaced by c-number procedures with respect to the characteristic function. When the Fourier transform of the characteristic function is taken, all procedures with respect to ξ, ξ^*, etc. are transformed into those with respect to u, u^*, v, v^*, D. Thus we obtain a differential equation for f. We give some details of this calculation in the following.

† The reader who is not familiar with the derivation of a classical Fokker–Planck equation is advised to read 4.12c.

Because the density matrix equation (317) consists of three parts referring to the electrons (atoms), the fields, and the interaction between electrons and field, we decompose $\mathrm{d}\chi/\mathrm{d}t$ correspondingly:

$$\left(\frac{\partial\chi}{\partial t}\right)_A = \sum_\mu \mathrm{tr}\left(O\frac{\partial\rho}{\partial t}\right)_{A,\mu}, \tag{355}$$

$$\left(\frac{\partial\chi}{\partial t}\right)_L = \mathrm{tr}\left\{O\left(\frac{\partial\rho}{\partial t}\right)_L\right\}, \tag{356}$$

$$\left(\frac{\partial\chi}{\partial t}\right)_{A-L} = \mathrm{tr}\left\{O\frac{-i}{\hbar}[H_{A-L},\rho]\right\}, \tag{357}$$

where the sum runs over all atoms, μ. Considering a single atom, we have

$$\mathrm{tr}\{O(a^\dagger{}_2 a_2 - a^\dagger{}_1 a_1)\,\rho\}.$$

We specialize O to a single atom by replacing

$$S_1{}^+, S_1{}^-, S_z \quad \text{by} \quad a_2{}^\dagger a_1, a_1{}^\dagger a_2; \quad (a_2{}^\dagger a_2 - a_1{}^\dagger a_1)/2$$

respectively and consider

$$\exp(i\xi^* a_2{}^\dagger a_1)\exp[i\zeta\tfrac{1}{2}(a_2{}^\dagger a_2 - a_1{}^\dagger a_1)]\exp(i\xi a_1{}^\dagger a_2)(a_2{}^\dagger a_2 - a_1{}^\dagger a_1). \tag{358}$$

We shift the last bracket through the last exponential function and calculate the commutator:

$$\exp(i\xi a_1{}^\dagger a_2)(a_2{}^\dagger a_2 - a_1{}^\dagger a_1) - (a_2{}^\dagger a_2 - a_1{}^\dagger a_1)\exp(i\xi a_1{}^\dagger a_2). \tag{358a}$$

We first expand

$$\exp(i\xi a_1{}^\dagger a_2) = 1 + i\xi a_1{}^\dagger a_2 + \tfrac{1}{2}(i\xi a_1{}^\dagger a_2)^2 + \ldots$$

which reduces on account of (225) and (224), to

$$\exp(i\xi a_1{}^\dagger a_2) = +1 i\xi a_1{}^\dagger a_2$$

Inserting this expression into (358a) and using again (225), we obtain

$$(358\mathrm{a}) = 2i\xi a_1{}^\dagger a_2,$$

which may be written in the form

$$2i\xi a_1{}^\dagger a_2 \exp(i\xi a_1{}^\dagger a_2).$$

Thus (358) =

$$\exp(i\xi^* a_2^\dagger a_1)\exp[i\zeta\tfrac{1}{2}(a_2^\dagger a_2 - a_1^\dagger a_1)]((a_2^\dagger a_2 - a_1^\dagger a_1) + 2i\xi a_1^\dagger a_2) \times \exp(i\xi a_1^\dagger a_2)$$

$$= 2\frac{\partial O}{i\partial\zeta} + 2\xi\frac{\partial O}{\partial\xi}. \tag{359}$$

Using (351) and (359) we find

$$\mathrm{tr}(O(a_2^\dagger a_2 - a_1^\dagger a_1)\,\rho) = 2\frac{\partial\chi}{i\partial\zeta} + 2\xi\frac{\partial\chi}{\partial\xi}. \tag{360}$$

When we now take the Fourier transform of the right-hand side of equation (360) we have

$$\int \exp(-i\xi v\ldots)\,\xi\frac{\partial\chi}{\partial\xi}\,\mathrm{d}\xi\ldots = i\frac{\partial}{\partial v}\int\exp(-i\xi v)\frac{\partial\chi}{\partial\xi}\,\mathrm{d}\xi\ldots$$

and, by partial integration

$$= i\frac{\partial}{\partial v}\left\{-\int\chi\frac{\partial}{\partial\xi}\exp(-i\xi v)\ldots\mathrm{d}\xi\ldots\right\}$$

$$= -\frac{\partial}{\partial v}\left(v\int\chi\exp(-i\xi v\ldots)\,\mathrm{d}\xi\ldots\right) = -\frac{\partial}{\partial v}(vf). \tag{361}$$

Consider as a further example, the expression

$$\exp(i\beta^* b^\dagger)\exp(i\beta b)\,b^\dagger b. \tag{362}$$

We shift $b^\dagger$ through $\exp(i\beta b)$

$$\exp(i\beta b)\,b^\dagger - b^\dagger\exp(i\beta b) = i\beta\exp(i\beta b). \tag{363}$$

Thus (362) gives

$$\exp(i\beta^* b^\dagger)(b^\dagger + i\beta)\exp(i\beta b)\,b \tag{364}$$

$$= \left(\frac{\partial^2}{\partial(i\beta)\,\partial(i\beta^*)} + i\beta\frac{\partial}{\partial(i\beta)}\right)\exp(i\beta^* b^\dagger)\exp(i\beta b) \tag{365}$$

and therefore

$$\mathrm{tr}(Ob^\dagger b) = \left(\frac{\partial^2}{\partial(i\beta)\,\partial(i\beta^*)} + i\beta\frac{\partial}{\partial(i\beta)}\right)\chi. \tag{366}$$

Taking the Fourier transform of (366) we can make similar transformations as above.

We hope that the above considerations elucidate the spirit of the derivation, so that we proceed to give the final equation for f:

$$\mathrm{d}f/\mathrm{d}t = Lf \tag{367}$$

where

$$L = L_A + L_{AL} + L_L$$

and

$$\begin{aligned} L_A = \frac{w_{12}}{2}\Bigg\{ & N\left[\exp\left(-2\frac{\partial}{\partial D}\right)-1\right]+N\exp\left(2\frac{\partial}{\partial D}\right)\frac{\partial^4}{\partial v^2\partial v^{*2}}+2N\frac{\partial^2}{\partial v\,\partial v^*} \\ & +\frac{\partial}{\partial v}\left[2\frac{\partial^2}{\partial v\,\partial v^*}+2\exp\left(-2\frac{\partial}{\partial D}\right)-1\right]v \\ & +\frac{\partial}{\partial v^*}\left[2\frac{\partial^2}{\partial v\,\partial v^*}+2\exp\left(-2\frac{\partial}{\partial D}\right)-1\right]v^* \\ & -2\left[\left[\exp\left(-2\frac{\partial}{\partial D}\right)-1\right]-\exp\left(2\frac{\partial}{\partial D}\right)\frac{\partial^4}{\partial_v{}^2\,\partial_v{}^{*2}}\right]\frac{D}{2}\Bigg\} \\ & +\frac{w_{21}}{2}\left\{N\left[\exp\left(2\frac{\partial}{\partial D}\right)-1\right]+\frac{\partial}{\partial v}v+\frac{\partial}{\partial v^*}v^*+2\left[\exp\left(2\frac{\partial}{\partial D}\right)-1\right]\frac{D}{2}\right\}, \end{aligned} \tag{368}$$

$$\begin{aligned} L_{AL} = -ig\Bigg\{ & \left[\exp\left(-2\frac{\partial}{\partial D}\right)v^*-\frac{\partial^2}{\partial v^2}v+\frac{\partial}{\partial v}D\right]u \\ & -\left[-\frac{\partial^2}{\partial v^{*2}}v^*+\exp\left(-2\frac{\partial}{\partial D}\right)v+\frac{\partial}{\partial v^*}D\right]u^* \\ & +\left[-\frac{\partial}{\partial u}+u^*\right]v-\left[-\frac{\partial}{\partial u^*}+u\right]v^*\Bigg\}, \end{aligned} \tag{369}$$

$$L_L = \kappa\left[\frac{\partial}{\partial u}u+\frac{\tilde{\upsilon}}{\partial u^*}u^*\right]+2\kappa n_{\mathrm{th}}\frac{\partial^2}{\partial u\,\partial u^*}. \tag{370}$$

4.11. THE CALCULATION OF MULTI-TIME CORRELATION FUNCTIONS BY MEANS OF THE DISTRIBUTION FUNCTION[64]

So far we have shown that we can derive a well defined equation for the distribution function (347). We have now to show how the properties of the

laser can be calculated by means of this function f. The laser properties, in particular its coherence properties, are determined by correlation functions of the form

$$\langle b^\dagger(t_1') \dots b^\dagger(t_m')\, b(t_n) \dots b(t_1)\rangle. \tag{371}$$

In (4.8) we have shown that (371) can be evaluated by the single atom density matrix. Because f is intimately connected with ρ, we start with the form (321).

The solution of eqn (324) defines $\rho(\alpha, \gamma)$, where the last term is explicitly given by (317). We define a generalized distribution function by

$$\begin{aligned} &f(u, u^*, v, v^*, D; \alpha(t), \gamma(t); t) \\ &= N' \int \dots \int \exp\left(-iv\xi - iv^*\xi^* - i\zeta \frac{D}{2} - iu\beta - iu^*\beta^*\right) \\ &\qquad X(\xi, \xi^*, \zeta, \beta, \beta^*; \alpha(t), \gamma(t))\, \mathrm{d}^2\xi\, \mathrm{d}\zeta\, \mathrm{d}^2\beta, \end{aligned} \tag{372}$$

where

$$\chi = \mathrm{tr}\{\exp(i\xi^* S^\dagger) \exp(i\zeta S_z) \exp(i\xi S^-) \exp(i\beta^* b^\dagger) \exp(i\beta b)\, \rho(\alpha, \gamma; t)\}. \tag{373}$$

Evidently the trace over $\rho(\alpha, \gamma)$ may be expressed by

$$\mathrm{tr}\{\rho(\alpha, \gamma, t)\} = \int f(u, u^*, v, v^*, D, \alpha, \gamma, t)\, \mathrm{d}^2u\, \mathrm{d}^2v\, \mathrm{d}D. \tag{374}$$

All steps which were done for the derivation of an equation for ρ may be simply repeated. We then find

$$\frac{\mathrm{d}f}{\mathrm{d}t} - Lf = -i\alpha(t)\, uf - i\gamma^*(t)\, u^* f. \tag{375}$$

We again introduce the decomposition (325) so that (375) becomes

$$\frac{\mathrm{d}f}{\mathrm{d}t} - Lf = 0, \tag{376}$$

for $t \neq t_i$ and additional jump conditions

$$f(t_i + 0) - f(t_i - 0) = -i\alpha_i\, uf(t_i - 0) + \text{corrections of order } \alpha^2. \tag{377}$$

We define a Green's function by

$$\frac{\mathrm{d}P}{\mathrm{d}t} - LP = \delta(t - t_1)\, \delta(v - v_1)\, \delta(u - u_1)\, \delta(D - D_1) \tag{378}$$

and use the notation

$$\begin{aligned} P(u_2, u_2^*, v_2, v_2^*, D_2; u_1, u_1^*, v_1, v_1^*, D_1; t_2, t_1) &= P(2, 1) \\ P(1, 0) &\equiv f(\alpha = 0; \gamma = 0). \end{aligned} \tag{379}$$

According to the jumps at time $t_1, t_2, t_3 \ldots$ we decompose the solution of eqn (376) into these time intervals, where we find for

$$t < t_1, \quad f = P(1, 0), \tag{380}$$

where $P(1,0)$ is the stationary state solution of eqn (376). For the next time interval we find

$$f = P(1, 0) + \int (-i\alpha_1 u_1 + i\gamma_1^* u_1^*) P(2, 1) P(1, 0)\, dV_1, \tag{381}$$

where

$$\mathrm{d}V_l = \mathrm{d}u_l\, \mathrm{d}u_l^*\, \mathrm{d}v_l\, \mathrm{d}v_l^*\, dD_l, \tag{382}$$

and for a time t after the last jump at t_n we obtain the expression

$$f = P(1,0) + \sum_{\nu=1}^{n} \underbrace{\int \ldots \int}_{\nu} \left\{ \prod_{\mu=1}^{\nu} P(\mu+1;\mu)(-i\alpha_\mu u_\mu + i\gamma_\mu^* u_\mu^*) \right\} P(1,0)\, \mathrm{d}V_1 \ldots \mathrm{d}V_\nu. \tag{383}$$

Because we must differentiate f finally with respect to all α's and γ^*'s respectively, we need only retain the term with $\nu = n$, so that we need only consider the equation

$$\left[\int f\, \mathrm{d}V\right]_n = \int_n \int \prod_{\mu=1}^{n} (-i\alpha_\mu u_\mu + i\gamma_\mu^* u_\mu^*) P(\mu, \mu-1)\, \mathrm{d}V_1 \ldots \mathrm{d}V_n, \tag{384}$$

where we have integrated both sides over $V_{n+1} = V$ and have used the relation

$$\int P(n+1, n)\, \mathrm{d}V_{n+1} = 1. \tag{385}$$

When we differentiate with respect to α and γ^* and make, if necessary, the transition to equal times (e.g. $t_i = t_{i+1}$) we find our final result in the following form

$$\langle (b^\dagger(t_1))^{\nu_1} (b^\dagger(t_2))^{\nu_2} \ldots (b^\dagger(t_n))^{\nu_n} (b(t_n))^{\mu_n} \ldots (b(t_1))^{\mu_1} \rangle$$
$$= \int_n \ldots \int u_n^{*\nu_n} u_n^{\mu_n} \ldots u_1^{*\nu_1} u_1^{\mu_1} \prod_{\mu=1}^{n} P(\mu, \mu-1)\, \mathrm{d}V_1 \ldots \mathrm{d}V_n. \tag{386}$$

Note that (386) is the most general formula, because by putting some of the μ's or ν's equal to zero, we may obtain all desired expectation values provided the operators are time ordered and in normal order.

4.12. REDUCTION OF THE GENERALIZED FOKKER–PLANCK EQUATION FOR TWO-LEVEL ATOMS AND FOR SINGLE-MODE ACTION

In this section we show how the exact generalized Fokker–Planck equation can be reduced step by step. In particular the adiabatic approximation is used which assumes that the relaxation times of the atomic system are much shorter than all other relevant times. This approximation holds to an excellent degree in the threshold region and above, up to rather high photon numbers. The resulting Fokker–Planck equation is valid for solid state, gas and semiconductor lasers. It is solved in Section 4.13, for the steady state. In 4.14 we shall represent the Fokker–Planck equation for multimode action near threshold and give its (nearly exact) stationary solution.

a. The first reduction: expansion in terms of powers of $N^{-\frac{1}{2}}$ *(*N *= number of atoms)*[65]

We introduce units

$$\tilde{u} = \frac{u}{\sqrt{n_{\text{thr}}}}, \qquad \tilde{v} = \frac{v}{v_{\text{thr}}}, \qquad \tilde{D} = \frac{D}{D_{\text{thr}}}, \tag{387}$$

where

$$n_{\text{thr}} = \delta_1 N^{\frac{1}{2}} \tag{388}$$

$$\delta_1 = \tfrac{1}{4}(\gamma_\perp \gamma_\parallel / \kappa(\kappa+\gamma_\perp))^{\frac{1}{2}}, \tag{389}$$

$$2\gamma_\perp = w_{12} + w_{21}, \qquad \gamma_\parallel = w_{12} + w_{21}, \tag{390}$$

$$v_{\text{thr}}{}^2 = \kappa^2 n_{\text{thr}}/g^2 = \delta_2 N^{3/2}/K, \tag{391}$$

$$K = g^2 N/\kappa\gamma_\perp = 1/\mathrm{d}_{\text{thr}}, \tag{392}$$

$$\delta_2 = \tfrac{1}{4}(\kappa\gamma_\parallel/\gamma_\perp(\kappa+\gamma_\perp))^{\frac{1}{2}}, \tag{393}$$

and

$$D_{\text{thr}} = N/K. \tag{394}$$

We consider K as fixed, but N to be very large. In the present context, n_{thr}, $v_{\text{thr}}{}^2$ and D_{thr} merely serve as abbreviations of the corresponding right-hand sides of eqns (388), (391) and (394). Later it will be shown, however, that these quantities are just the values of the photon number, of the absolute square of the total atomic dipole moment, and of the total inversion *at threshold.* The threshold inversion of the single atom is d_{thr}. Taking into

account terms of the order one and $N^{-\frac{1}{2}}$, we find

$$L_A = (w_{21}-w_{12})\,K\frac{\partial}{\partial\tilde{D}}+\gamma_\perp\left(\frac{\partial}{\partial\tilde{v}}\,\tilde{v}+\frac{\partial}{\partial\tilde{v}^*}\,\tilde{v}^*\right)+\gamma_\parallel\frac{\partial}{\partial\tilde{D}}\,\tilde{D}+N^{-\frac{1}{2}}\frac{w_{12}\,K}{\delta_2}\,\frac{\partial^2}{\partial\tilde{v}\,\partial\tilde{v}^*}. \tag{395}$$

$$L_{A-F} = -ig\Big\{-2\frac{\partial}{\partial\tilde{D}}\,\tilde{v}^*\,\tilde{u}\sqrt{(\delta_1\delta_2\,K)}$$

$$+\frac{\partial}{\partial\tilde{v}}\,\tilde{D}\tilde{u}\,\sqrt{(\delta_1/\delta_2)}\,\frac{1}{\sqrt{K}}\,N^{\frac{1}{2}}-\frac{\partial}{\partial\tilde{u}}\,\tilde{v}\,\sqrt{(\delta_2/\delta_1)}\,\frac{1}{\sqrt{K}}\,N^{\frac{1}{2}}-\text{c.c.}\Big\} \tag{396}$$

Since $g \sim N^{-\frac{1}{2}}$ for fixed K, (392), we have neglected all terms in the curly bracket which are proportional to $N^{-\frac{1}{2}}, N^{-1}, \ldots$. In order to obtain a concise formula, we replace the complex variables u and v by two-dimensional vectors thus

$$(u, u^*) \to \vec{u} \qquad -i(v, v^*) \to \vec{v}.$$

Also, we put

$$D_0 = N\frac{w_{12}-w_{21}}{w_{12}+w_{21}}. \tag{397}$$

After a short transformation we obtain

$$\frac{\partial f}{\partial t}+\nabla_{\mathbf{u}}\{(-\kappa\mathbf{u}+g\mathbf{v})f\}+\Delta_{\mathbf{v}}\{(-\gamma_\perp\,\mathbf{v}+gD\mathbf{u})f\}+\frac{\partial}{\partial D}\{[\gamma_\parallel(D_0-D)\,-4g\mathbf{uv}]f\}$$

$$=\frac{\kappa}{2}\,n_{th}\,\Delta_{\mathbf{u}}f+\tfrac{1}{4}Nw_{12}\,\Delta_{\mathbf{v}}f+\gamma_\parallel\,N\,\frac{\partial^2 f}{\partial D^2}. \tag{398}$$

An equation of this form was first obtained by Risken *et al.*[45] There is only a slight difference between their equation and eqn (398). They used symmetrized diffusion coefficients and found $n_{\text{thr}}+\frac{1}{2}$ instead of n_{thr} and $\frac{1}{4}N(w_{21}+w_{12})/2$ instead of w_{12} in the first two terms. A detailed analysis shows that both forms lead to the same results, as long as higher derivatives are neglected.

b. Adiabatic elimination of the atomic variables

With respect to the statistical properties of the laser field, the distribution function f contains more information than is needed, because it also contains the atomic variables. Thus it is desirable to eliminate these variables, so that a function $W(u, u^*)$ of the field variables alone can be determined. If the relaxation times of the atomic system are short compared to all other times of

the systems, the atomic variables (i.e. the dipole moment and the inversion) follow the motion of the field adiabatically. Under this assumption the atomic variable can be easily eliminated and, to do this, two ways are known:

(1) direct elimination within the Fokker–Planck equation;[66]

(2) elimination via the Langevin equations.[67]

We present the second procedure because it is easier to follow.

The Fokker–Planck equation (398) possesses the following associated Langevin equations for the complex light amplitude u, the total dipole moment v, and the inversion D:

$$\left(\frac{\mathrm{d}}{\mathrm{d}t}+\kappa\right) u+igv = \Gamma_u, \tag{399}$$

$$\left(\frac{\mathrm{d}}{\mathrm{d}t}+\gamma_\perp\right) v-iguD = i\Gamma_v, \tag{400}$$

$$\left(\frac{\mathrm{d}}{\mathrm{d}t}+\gamma_\parallel\right)(D-D_0)+2ig(v^*u-vu^*) = \Gamma_D. \tag{401}$$

Note that these equations are now *classical* with *classical* random forces. The diffusion constants are given by

$$Q_j = \frac{1}{4\Gamma}\int_0^T\int_0^T \langle \Gamma_j(t_1)\,\Gamma_j^*(t_2)\rangle\,\mathrm{d}t_1\,\mathrm{d}t_2. \tag{402}$$

A comparison with the RSW–Fokker–Planck equation[67] (in the symmetrized form) yields

$$Q_u = \frac{\kappa}{2}(n_{\mathrm{th}}+\tfrac{1}{2}), \tag{403}$$

$$Q_v = \tfrac{1}{4}N\gamma_\perp = \tfrac{1}{4}(N_1+N_2)\,\gamma_\perp = \tfrac{1}{4}N\frac{w_{21}+w_{12}}{2}. \tag{404}$$

Had we used the Fokker–Planck equation (398) instead, the result would read

$$Q_u = \kappa n_{\mathrm{th}}/2, \tag{405}$$

$$Q_v = Nw_{12}/4. \tag{406}$$

We shall see below that the results are not affected by the difference between (403), (404) and (405), (406) so that the corresponding Fokker–Planck equations are equivalent. We now eliminate the atomic variables. In accordance with the adiabatic assumption we neglect $\mathrm{d}/\mathrm{d}t$ compared to $\gamma_\perp$ and $\gamma_\parallel$.

Neglecting Γ_D too, we obtain

$$D = D_0 - \frac{4g^2}{\gamma_\parallel \gamma_\perp} D_0 \, uu^*. \tag{407}$$

Inserting (407) into (400) and again neglecting $\mathrm{d}/\mathrm{d}t$ we obtain the dipole moment as a function of the field alone. Introducing the corresponding expression into the field equation (399) we find

$$\frac{\mathrm{d}}{\mathrm{d}t} u - \beta(\bar{n} - u^* u)\, u = \Gamma, \tag{408}$$†

where

$$\beta = 4 \frac{\kappa^2 \gamma_\perp}{\gamma_\parallel (\kappa + \gamma_\perp)(N_2 - N_1)_{\mathrm{thr}}}, \tag{409}$$

$$\bar{n} = \tilde{d} = \frac{\gamma_\parallel}{4\kappa} (D_0 - D_{\mathrm{thr}}), \tag{410}$$

$$D_{\mathrm{thr}} \equiv (N_2 - N_1)_{\mathrm{thr}} = \kappa \gamma_\perp / g^2, \tag{411}$$

$$\Gamma = \frac{\gamma_\perp}{\kappa + \gamma_\perp} \Gamma_u + \frac{2}{\kappa + \gamma_\perp} \Gamma_v. \tag{412}$$

c. Classical derivation of Fokker–Planck equation‡

In the foregoing chapters we have shown how the Fokker–Planck equation for a laser near threshold can be derived step by step, starting from the quantum mechanical density matrix equations. The resulting equation can also be obtained in a more heuristic way by interpreting the quantum mechanical van der Pol equation (267) as a classical equation. We briefly remind the reader by means of an example how the Fokker–Planck equation is derived for a Markoff-process. Consider the equation of motion of a classical Brownian particle, which is moving e.g. in a liquid or gas. Its velocity χ changes due to a friction force $-\gamma\chi$ and a randomly fluctuating (Langevin) force $F(t)$. We assume that the ensemble average over $F(t)$ vanishes: $\langle F(t) \rangle$, and that the reservoir i.e. the surrounding gas causing the fluctuations of $F(t)$, has a very short memory, so that

$$\langle F(t)\, F(t') \rangle \sim \delta(t - t') \tag{413}$$

† This equation is slightly more general than eqn (267) in that we have now $\kappa + \gamma_\perp$ instead of $\gamma_\perp$ ($\equiv \gamma$).

‡ Readers who are familiar with the derivation of a classical Fokker–Planck equation may proceed directly to 4.12d.

We now want to derive an equation as to what the probability is of finding the particle with a velocity χ at a given time. For each realization of the motion, i.e. for a given sequence of pushes described by F, we obtain a definite "path". The probability of finding the particle of a velocity χ is then

$$P(\chi, t) = \delta(\chi - \chi_1(t)). \tag{414}$$

If the "gedanken" experiment is repeated, we would find another path realized:

$$\left.\begin{aligned} P(\chi, t) &= \delta(\chi - \chi_2(t)), \\ P(\chi, t) &= \delta(\chi - \chi_3(t)). \\ &\vdots \end{aligned}\right\} \tag{415}$$

When the experiment is repeated often, we can define an average value of P by

$$f(\chi, t) = \langle P(\chi, t)\rangle = \frac{\sum_i w_i \delta(\chi - \chi_i(t))}{\sum_i w_i} \equiv \langle \delta(\chi - \chi(t))\rangle, \tag{416}$$

where w_i is the frequency (or mathematically speaking, the probability) of realization of the special path i. In order to derive an equation for f we form

$$\begin{aligned} \Delta f(\chi, t) &\equiv f(\chi, t+\Delta t) - f(\chi, t) \\ &= \langle \delta(\chi - \chi(t+\Delta t))\rangle - \langle \delta(\chi - \chi(t))\rangle. \end{aligned} \tag{417}$$

Using

$$\chi(t+\Delta t) = \chi(t) + \Delta\chi(t), \tag{418}$$

we expand the right-hand side of eqn (417):

$$\Delta f(\chi, t) = \left\langle \left(-\frac{\mathrm{d}}{\mathrm{d}\chi} \delta(\chi - \chi(t)) \right) \Delta\chi(t) \right\rangle + \tfrac{1}{2} \left\langle \frac{\mathrm{d}^2}{\mathrm{d}\chi^2} \delta(\chi - \chi(t)) (\Delta\chi(t))^2 \right\rangle. \tag{419}$$

We now use the Langevin equation which describes the motion of the Brownian particle:

$$\chi(t) = -\gamma\chi(t) + F(t), \tag{420}$$

from which we find

$$\Delta\chi = -\gamma\chi\Delta t + \Delta F, \tag{421}$$

where Δt is so small that χ has changed only little, but so large that ΔF contains many pushes. We use $\langle \Delta F\rangle = 0$ and find further on account of (413):

$$\langle \Delta F(t)\, \Delta F(t)\rangle = 2Q \,.\, \Delta t \tag{422}$$

i.e. a proportionality to Δt.

We now treat

$$\left\langle \frac{\mathrm{d}}{\mathrm{d}\chi}\delta(\chi-\chi(t))\,\Delta\chi(t)\right\rangle. \tag{423}$$

Because χ does not depend on the averaging procedures, we may write

$$(423) = \frac{\mathrm{d}}{\mathrm{d}\chi}\langle\delta(\chi-\chi(t))\,\Delta\chi(t)\rangle. \tag{424}$$

Replacing $\Delta\chi(t)$ according to (421) we find

$$(424) = \frac{\mathrm{d}}{\mathrm{d}\chi}\langle\delta(\chi-\chi(t))(\chi(t)\,\Delta t(-\gamma)+\Delta F(t))\rangle. \tag{425}$$

Due to the δ-function we may replace $\chi(t)$ by χ; due to the Markoff assumption we may split the term containing $\Delta F(t)$ into a product of averages, so that we have finally:

$$\left\langle \frac{\mathrm{d}}{\mathrm{d}\chi}\delta(\chi-\chi(t))\,\Delta\chi(t)\right\rangle = \frac{\mathrm{d}}{\mathrm{d}\chi}\{\langle\delta(\chi-\chi(t))\rangle\langle(-\Delta t\gamma\chi)\rangle+\langle\delta(\chi-\chi(t))\rangle\langle\Delta F(t)\rangle\}, \tag{426}$$

where $$\langle\Delta F(t)\rangle = 0.$$

We treat the second term in (419) similarly. Keeping terms $\sim\Delta t$ and dropping those $\sim(\Delta t)^2$, we find

$$\left\langle \frac{\mathrm{d}^2}{\mathrm{d}\chi^2}\delta(\chi-\chi(t))(\Delta\chi)^2\right\rangle \approx \frac{\mathrm{d}^2}{\mathrm{d}\chi^2}\langle\delta(\chi-\chi(t))\,.\,2Q\,.\,\Delta t. \tag{427}$$

Inserting (425) and (427) into (419), dividing the resulting equation by Δt and letting

$$\lim_{\Delta t\to 0}\frac{\Delta f}{\Delta t} = \frac{\mathrm{d}f}{\mathrm{d}t},$$

we obtain the Fokker–Planck equation

$$\frac{\mathrm{d}f}{\mathrm{d}t} = -\frac{\mathrm{d}}{\mathrm{d}\chi}(-\gamma\chi f(\chi,t))+Q\frac{\mathrm{d}^2}{\mathrm{d}\chi^2}f(\chi,t). \tag{428}$$

On account of the Markoff assumption, no higher derivatives occur. The derivation goes through the same way if the friction force $-\gamma\chi$ is replaced by a nonlinear force $K(\chi)$, e.g. of the form

$$K = +\beta\chi(d-\chi^2). \tag{429}$$

The derivation is easily extended to e.g. three dimensions or more variables. Let us return to the laser, described by the Langevin equation (408) We decompose the complex amplitude b^*:

$$b^* = r\exp(i\phi) = \chi + iy \tag{430}$$

and replace f by $W(\chi, y)$ or $W(r, \phi)$. The Fokker–Planck equation for a particle moving in two dimensions under a friction force

$$\left.\begin{aligned} K_\chi &= +\beta\chi(\tilde{d} - (\chi^2 + y^2)) \\ K_y &= +\beta y(\tilde{d} - (\chi^2 + y^2)) \end{aligned}\right\} \tag{431}$$

is given in analogy to (428):

$$\frac{\mathrm{d}f}{\mathrm{d}t} = -\frac{\mathrm{d}}{\mathrm{d}\chi}\{\beta\chi(\tilde{d} - (\chi^2 + y^2))f\} - \frac{\mathrm{d}}{\mathrm{d}y}\{\beta y(\tilde{d} - (\chi^2 + y^2))f\} + Q\left(\frac{\mathrm{d}^2}{\mathrm{d}\chi^2} + \frac{\mathrm{d}^2}{\mathrm{d}y^2}\right)f. \tag{432}$$

d. The Fokker–Planck equation

We write the complex field amplitude $u\,(\equiv b^*)$, in polar coordinates: $u = r\exp(i\phi)$. The Fokker–Planck equation belonging to the *classical van der Pol equation* (in the rotating wave approximation (408) or (267)) reads[68]

$$\frac{\partial W}{\partial t} + \beta\frac{1}{r}\frac{\partial}{\partial r}\{(\tilde{d} - r^2)\, r^2\, W\} = Q\left\{\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial W}{\partial r}\right) + \frac{1}{r^2}\frac{\partial^2 W}{\partial \phi^2}\right\}. \tag{433}$$

The diffusion constant Q is given by

$$Q = \tfrac{1}{4}\frac{1}{T}\int_0^T\int_0^T \langle \Gamma(t_1)\,\Gamma^*(t_2)\rangle\,\mathrm{d}t_1\,\mathrm{d}t_2. \tag{434}$$

Using the form (412) of Γ and the fact that Γ_u and Γ_v are uncorrelated, we obtain

$$Q = \left(\frac{\gamma_\perp}{\kappa + \gamma_\perp}\right)^2 Q_u + \frac{g^2}{(\kappa + \gamma_\perp)^2}\, Q_v. \tag{435}$$

Inserting (403) and (404) into (435) we find

$$Q = \left(\frac{\gamma_\perp}{\kappa + \gamma_\perp}\right)^2 \frac{\kappa}{2}(n_{\mathrm{th}} + \tfrac{1}{2}) + \frac{g^2}{(\kappa + \gamma_\perp)^2}\,\tfrac{1}{4}N\gamma_\perp. \tag{436}$$

We now eliminate the coupling constant using the threshold condition

$$(N_2 - N_1)_{\mathrm{thr}} = \kappa\gamma_\perp/g^2. \tag{437}$$

After re-arranging (436) we obtain, finally,

$$Q = \frac{\kappa}{2}\left[\frac{\gamma_\perp}{\kappa+\gamma_\perp}\right]^2\left[n_{\text{th}}+\tfrac{1}{2}+\tfrac{1}{2}\frac{(N_2+N_1)_{\text{thr}}}{(N_2-N_1)_{\text{thr}}}\right]$$
$$= \frac{\kappa}{2}\left[\frac{\gamma_\perp}{\kappa+\gamma_\perp}\right]^2\left[n_{\text{th}}+\frac{N_{2\text{thr}}}{(N_2-N_1)_{\text{thr}}}\right]. \tag{438}$$

(Note that $N = N_2+N_1 = (N_2+N_1)_{\text{thr}}$).

Let us now discuss the diffusion coefficient using (405) and (406):

$$Q = \left(\frac{\gamma_\perp}{\kappa+\gamma_\perp}\right)^2\frac{\kappa}{2}n_{\text{th}}+\frac{\kappa\gamma_\perp}{(\kappa+\gamma_\perp)^2}\frac{1}{(N_2-N_{1\text{thr}})}\tfrac{1}{4}Nw_{12}, \tag{439}$$

where use of (437) has been made again. Because $N_2 = (w_{12}/(w_{12}+w_{21}))\,N$ at threshold, (439) and (438) are indeed equivalent, provided that the photon numbers are not too large.

4.13. SOLUTION OF THE REDUCED FOKKER–PLANCK EQUATION

a. Steady state solution

First we introduce normalized coordinates:

$$\hat{r} = \sqrt[4]{(\beta/Q)}\,r; \qquad \hat{t} = \sqrt{(\beta Q)}\,t; \qquad \tilde{d} = \sqrt{(Q/\beta)}\,a. \tag{440}$$

Using these, the Fokker–Planck equation then takes the form

$$\frac{\partial W}{\partial \hat{t}}+\frac{1}{\hat{r}}\frac{\partial}{\partial \hat{r}}\{(a-\hat{r}^2)\hat{r}^2W\} = \frac{1}{\hat{r}}\frac{\partial}{\partial \hat{r}}\left(\hat{r}\frac{\partial W}{\partial \hat{r}}\right)+\frac{1}{\hat{r}^2}\frac{\partial^2 W}{\partial \phi^2}. \tag{441}$$

Its stationary solution is given by[68]

$$W(\hat{r}) = \frac{N}{2\pi}\exp\left[-\frac{\hat{r}^4}{4}+a\frac{\hat{r}^2}{2}\right], \qquad \frac{1}{N} = \int_0^\infty \hat{r}\exp\left[-\frac{\hat{r}^4}{4}+a\frac{\hat{r}^2}{2}\right]\mathrm{d}\hat{r}. \tag{442}$$

The distribution function (442) is a continuous function of $\hat{r}$ (or r). In order to compare it with the distribution, $p(n, T)$, of the discrete photon numbers, n, which are measured in the time-interval T, a correction derived by Mandel must be added. If T is small compared to the relaxation times of the van der Pol oscillator, $p(n, T)$ is given by

$$P(n, T) = \int_0^\infty \frac{(\hat{\alpha}\hat{I})^n}{n!}\exp(-\hat{\alpha}\hat{I})\,\tilde{W}(\hat{I})\,\mathrm{d}\hat{I}, \tag{443}$$

where†

$$\hat{I} = \hat{r}^2, \qquad \hat{\alpha} = \alpha\sqrt{(Q/\beta)}\,.\,T.$$

On account of $\tilde{W}(\hat{I}) = \pi W(\hat{r})$ and using the explicit form of $W(\hat{r})$ (442) we find

$$P(n, T) = \frac{\hat{\alpha}^n}{n!}\,\frac{F_n(a-2\hat{\alpha})}{F_0(a)}, \tag{444}$$

where $F_n(a)$ is given by

$$F_n(a) = \int_0^\infty \hat{I}^n \exp\left[-\frac{\hat{I}^2}{4} + a\,\frac{\hat{I}}{2}\right] \mathrm{d}\hat{I}. \tag{445}$$

For a large average number of photons, i.e. for large $\hat{\alpha}$ we obtain a (nearly) continuous distribution

$$P(n, T) = \frac{1}{\hat{\alpha}F_0(a)} \exp\left\{-\frac{1}{4}\left(\frac{n}{\hat{\alpha}}\right)^2 + \frac{1}{2}\,a\,\frac{n}{\hat{\alpha}}\right\}. \tag{446}$$

b. Non-stationary solutions

In order to obtain *correlation functions*, the *non-stationary solutions* of the Fokker–Planck equations must be used. Since general analytical expressions are not known, approximations must be used or computer solutions must be sought. There are some exact relations, however. If we denote the Green's function belonging to (441) by

$$F(\hat{r}, \phi; \hat{r}', \phi', \hat{\tau}),$$

the correlation function of the intensity fluctuations is given by[69]

$$\begin{aligned} K(a, \hat{\tau}) &\equiv \langle(\hat{r}^2(\hat{t}+\hat{\tau}) - \langle\hat{r}^2\rangle)(\hat{r}^2(\hat{t}) - \langle\hat{r}^2\rangle)\rangle \\ &\equiv \int\int\int\int \hat{r}\,\mathrm{d}\hat{r}\,\hat{r}'\,\mathrm{d}\hat{r}'\,\mathrm{d}\phi\,\mathrm{d}\phi'(\hat{r}^2 - \langle\hat{r}^2\rangle)(\hat{r}'^2 - \langle\hat{r}'^2\rangle)\,F(\hat{r}, \phi; \hat{r}', \phi', \hat{\tau}). \end{aligned} \tag{447}$$

Expressing F by its decomposition into eigenfunctions, one finds

$$K(a, \hat{\tau}) = K(a, 0) \sum_{m=1}^{\infty} M_m \exp(-\lambda_{0m}\hat{\tau}). \tag{448}$$

The M_m's are certain moments of $\hat{r}$ with respect to the eigenfunctions, λ_{0m}'s are the corresponding eigenvalues.

The spectrum $S(a, \hat{\omega})$ of the intensity fluctuations is given by the Fourier transform of the correlation function (448)‡

† α is a proportionality factor which connects the probability of measuring a single photon in the infinitesimal time interval $\mathrm{d}t$ with intensity $I = P(1, \mathrm{d}t) = \alpha I\,\mathrm{d}t$.

‡ The use of $\hat{\omega}$ instead of ω indicates that we are using reduced units.

$$S(a,\hat{\omega}) = K(a,0)\sum_{m=1}^{\infty} M_m \frac{\lambda_{0m}}{\hat{\omega}^2+\lambda^2{}_{0m}}. \tag{449}$$

Although it is a sum of Lorentzian lines of widths λ_{0m}, the spectrum $S(a,\hat{\omega})$ may be well approximated by an "effective" Lorentzian curve[70]

$$S_{\text{eff}}(a,\hat{\omega}) = K(a,0)\frac{\lambda_{\text{eff}}}{\hat{\omega}^2+\lambda^2{}_{\text{eff}}}$$

with

$$\frac{1}{\lambda_{\text{eff}}} = \sum_{m=1}^{\infty}\frac{M_m}{\lambda_{0m}}, \tag{450}$$

which has the same area and the same maximum value. This effective width λ_{eff} is, however, about 25% larger than λ_{01} for $a \approx 5$. The eigenvalues and matrix elements were calculated numerically by Risken and Vollmer[70] for the threshold region $-10 \leqslant a \leqslant 10$. The results of Risken and Vollmer were experimentally checked by a number of authors and excellent agreement was found (see the chapter by E. R. Pike in this volume, where the relevant references are given).

4.14. THE FOKKER–PLANCK EQUATION FOR MULTIMODE ACTION NEAR THRESHOLD

a. Stationary solution

In Section 4.10 we have derived an exact generalized Fokker–Planck equation for a single mode and a system of two-level atoms. In a region not too far above threshold we have reduced that equation to an ordinary Fokker–Planck equation for the field variables alone. The whole procedure can be extended to multimode laser action with multi-level atoms. We do not intend to repeat all the steps in detail, because the equations are very lengthy. We rather want to describe how to derive this final equation be merely looking at some of the intermediate results of our single-mode analysis.

The first important result was that the Fokker–Planck equation, after the first reduction, can be interpreted as being connected with a set of classical Langevin equations. If we remember the results of Section 3 on the semi-classical treatment of the laser equations, we find by comparison, that the classical Langevin equations coincide with the *semi-classical* laser equations (if the fluctuating forces are dropped). Because the drift coefficients are identical to the right-hand sides of these Langevin equations (without fluctuating forces), we can immediately use the corresponding expressions of the semiclassical analysis. Furthermore, we have seen that the subsequent adiabatic elimination can best be done in the framework of the Langevin equations. This elimination procedure has been performed in Section 3 for

multimode action, so that we can use the results of this chapter for the explicit form of the drift coefficients. Finally it is obvious from our above analysis that the diffusion coefficients can be found from fluctuating forces of the quantum mechanical Langevin equations. It can be shown that the diffusion coefficients Q_{ij} vanish for different modes, $i.j$. The proof of this simply uses the fact that the thermal heatbaths of the modes are uncorrelated and that the modes are orthogonal with respect to their spatial dependence.

With these considerations, it is quite simple to write down the Fokker–Planck equation for multimode laser action in homogeneously or inhomogeneously broadened lasers. Denoting the complex field amplitudes of the modes λ by u_λ, u_λ^*, we obtain

$$\frac{\mathrm{d}}{\mathrm{d}t} W(u_1, \ldots, u_M) = \Big\{ -\sum_\lambda \frac{\partial}{\partial u_\lambda} A_\lambda(u_1, \ldots, u_M) - \sum_\lambda \frac{\partial}{\partial u_\lambda^*} A_\lambda^*(u_1, \ldots, u_M) + \sum_\lambda Q_\lambda \frac{\partial^2}{\partial u_\lambda \, \partial u_\lambda^*} \Big\} W(u_1, \ldots, u_M), \tag{451}$$

where

$$\begin{aligned} A_\lambda^*(u_1, \ldots, u_M) = {} & (i\omega_\lambda - \kappa_\lambda) u_\lambda^* - u_\lambda^* \sum_\mu d_{0\mu} |g_{\mu\lambda}|^2 \frac{i}{\Omega_\lambda - \nu_\mu - i\gamma_\perp} \\ & + 2 \sum_{\mu\lambda_1\lambda_2\lambda_3} d_{0\mu} \, g_{\mu\lambda}^* \, g_{\mu\lambda_1}^* \, g_{\mu\lambda_2} \, g_{\mu\lambda_3} \, u_{\lambda_1} \, u_{\lambda_2}^* \, u_{\lambda_3}^* \\ & \times \left\{ \left(\frac{1}{T} + i(\Omega_{\lambda_2} - \Omega_{\lambda_1}) \right) (\Omega_{\lambda_2} + \Omega_{\lambda_3} - \Omega_{\lambda_1} - \nu_\mu - i\gamma_\perp) \right\}^{-1} \\ & \times \frac{\Omega_{\lambda_1} - \Omega_{\lambda_2} + 2i\gamma_\perp}{(\Omega_{\lambda_2} - \nu_\mu - i\gamma_\perp)(\Omega_{\lambda_1} - \nu_\mu + i\gamma_\perp)}. \end{aligned} \tag{452}$$

In these equations ω_λ and κ_λ are mode frequency and halfwidth in the unloaded cavity; Ω_λ is the mode frequency in the loaded cavity; $d_{0\mu}$ is the unsaturated inversion of atom μ; $g_{\mu\lambda}$ is the coupling constant; ν_μ is the frequency at line centre of atom μ; $\gamma_\perp$ is the homogeneous atomic width; and T is the longitudinal relaxation time. Further, we have for the diffusion coefficients,

$$Q_\lambda = \frac{1}{8} \left\{ \sum_\mu \frac{2\gamma_\perp (N_2 + N_1)_\mu}{\gamma_\perp^2 + (\nu_\mu - \Omega_\lambda)^2} |g_{\mu\lambda}|^2 + 2\kappa(2n_{\mathrm{th}} + 1) \right\}. \tag{453}$$

Before giving the (nearly exact) solution of (451) with (452) and (453) we present a theorem.

b. Theorem on the exact stationary solution of a Fokker–Planck equation[71]

We assume that the drift and diffusion coefficients of the Fokker–Planck equation

$$\frac{\partial W}{\partial t} = \left\{ \sum_j \left(\frac{\partial}{\partial u_j} C_j + \frac{\partial}{\partial u_j^*} \tilde{C}_j \right) + \sum_{kj} Q_{kj} \frac{\partial^2}{\partial u_k \partial u_j^*} \right\} W \tag{454}$$

have the following form

$$C_j = \frac{\partial B}{\partial u_j^*} + \mathscr{T}_j^{(1)}, \tag{455}$$

$$\tilde{C}_j = \frac{\partial B}{\partial u_j} + \mathscr{T}_j^{(2)}, \tag{456}$$

$$Q_{kj} = \delta_{kj} Q. \tag{457}$$

We assume also that

$$\sum_j \left(\frac{\partial B}{\partial u_j} \mathscr{T}_j^{(1)} + \frac{\partial B}{\partial u_j^*} \mathscr{T}_j^{(2)} \right) = 0, \tag{458}$$

$$\sum_j \left(\frac{\partial \mathscr{T}_j^{(1)}}{\partial u_j} + \frac{\partial \mathscr{T}_j^{(2)}}{\partial u_j^*} \right) = 0. \tag{459}$$

The theorem states that the stationary solution of the Fokker–Planck equation (454) is given by

$$W = N \exp(-2B/Q), \tag{460}$$

where N is the normalization constant. It is implicitly assumed that W is normalizable which is always the case in laser systems because of the gain saturation.

c. Nearly exact solution of (451)

Because modes near the atomic line centre have the highest gain, we confine our analysis to modes for which Q_λ is nearly independent of λ, so that the condition (457) is fulfilled. Whether A^* and A (452) fulfil the condition (455), (456), (458) and (459), requires a detailed and lengthy analysis, so that we represent only the results for the important example of normal multimode action.

We assume that the frequency spacing $\Delta\Omega = \Omega_{\lambda_2} - \Omega_{\lambda_1}$ is large compared to $1/T$. The important contributions to (452) then stem from those terms in the sum where $\lambda_1 = \lambda_2$. If we further neglect combination tones, the sum over λ_3

reduces to a single term: $\lambda_3 = \lambda$. Under these assumptions eqn (451) possesses the following exact solution,

$$W = N \exp\left\{ -\frac{2}{Q}(B^{(1)}+B^{(2)}+B^{(3)}) \right\}, \tag{461}$$

where

$$B^{(1)} = \sum_{\lambda} \kappa_\lambda u_\lambda^* u_\lambda, \tag{462}$$

is connected with the cavity losses,

$$B^{(2)} = \sum_{\lambda} \Delta\kappa_\lambda u_\lambda^* u_\lambda \tag{463}$$

with

$$\Delta\kappa_\lambda = -\sum_{\mu} d_{0\mu}|g_{\mu\lambda}|^2 \frac{\gamma_\perp}{(\Omega_\lambda - \nu_\mu)^2 + \gamma_\perp{}^2} \tag{464}$$

is connected with the unsaturated gain, and

$$B^{(3)} = 2\gamma_\perp{}^2 T \sum_{\lambda\lambda_1\mu} |u_\lambda|^2 |u_{\lambda_1}|^2 d_{0\mu}|g_{\mu\lambda}|^2 |g_{\mu\lambda_1}|^2 \frac{1}{(\Omega_\lambda - \nu_\mu)^2 + \gamma_\perp{}^2} \cdot \frac{1}{(\Omega_{\lambda_1} - \nu_\mu)^2 + \gamma_\perp{}^2} \tag{465}$$

is connected with the gain saturation (including mode interaction). (If (461) with (462)–(465) is specialized to a single mode, the result (442) follows.)

5. The General Theory of Dissipation and Fluctuations of Quantum Systems Far From Thermal Equilibrium

5.1. The quantum system

We adopt the method of projection operators. We assume that a system can be described by a set of quantum states which are distinguished by a quantum number l. In the following we always assume that l stands for the complete set which characterizes the quantum states of the system and we shall only use the property that the projection operators P_{ik} form an algebraic ring with the property

$$P_{ik} P_{lm} = P_{im} \delta_{kl}. \tag{466}$$

By means of these projection operators it is possible to decompose every operator Ω into

$$\Omega = \sum_{lm} \langle l|\Omega|m\rangle P_{lm}. \tag{467}$$

For a N level atom with a single electron, P_{lm} can be written in terms of creation and annihilation operators, thus $P_{lm} = a_l^\dagger a_m$.

5.2. THE GENERAL QUANTUM MECHANICAL LANGEVIN EQUATIONS[72]

We assume that transition rates w and damping constants γ are given, either by a calculation from first principles or, what is of great practical importance, phenomenologically. We require further that the fluctuating forces have two properties:

1. they have a short memory, so that their correlation function is proportional to a δ-function of the time difference (or, in other words, they have Markoff character).

2. they are determined in such a way, that the relation for projection operators holds, at least if the average is taken

$$\langle P_{ik} P_{lm} \rangle = \delta_{kl} \langle P_{im} \rangle, \tag{468}$$

where the P's are the time-dependent operators under the influence of the full Heisenberg operator, which comprises also the effect of the heatbaths. We show that the fluctuating forces can then be determined uniquely and explicitly by a rather simple formula (see (486) and (490)). They secure also the quantum mechanical consistency of the equations of motion for the operators P_{ik}. We start with the averaged equations which we write in the general form

$$\frac{\mathrm{d}}{\mathrm{d}t} \langle P_{i_1 i_2} \rangle = \sum_{j_1 j_2} \langle M_{i_1 i_2 ; j_1 j_2} P_{j_1 j_2} \rangle \tag{469}$$

The number of levels 1, 2, . . . may be arbitrary. Denoting the vector formed of $P_{i_1 i_2}$ by A and the matrix formed of $M_{i_1 i_2 ; j_1 j_2}$ by M (note that $i_1 i_2$ counts as a single index) we can represent eqn (469) as

$$\frac{\mathrm{d}}{\mathrm{d}t} \langle A \rangle = \langle MA \rangle. \tag{470}$$

In it, M contains the transition probabilities (especially the losses), the non-diagonal phase-destroying elements produced by the heatbath, and the "coherent" driving field. Because these driving fields, if treated quantum mechanically, may still depend in an implicit way on the heatbath, the average over the heatbath has to comprise both M and A. We now go one step back and consider the motion of the unaveraged operators A by adding fluctuating driving forces, Γ.

$$\dot{A} = MA + \Gamma. \tag{471}$$

The driving forces can have any form and can especially still depend on P_{ik} for instance, in the following form

$$\Gamma(t) = L(t)A + N(t), \tag{472}$$

where L and N do not depend on A.

In order to come back from (471) to (470) we must assume

$$\langle \Gamma \rangle = 0. \tag{473}$$

It should be noted that we have to impose as an additional condition, the conservation of the total probability, expressed by

$$\sum_{i=1}^{N} P_{ii} = 1. \tag{474}$$

The formal solution of (471) can be written as an integral representation

$$A = \int^{t} K(t, \tau)\, \Gamma(\tau)\, \mathrm{d}\tau + A_n, \tag{475}$$

where A_n is a solution of the homogeneous part of (471). We postulate in the well known way that the kernel has the property

$$K(t, t) = E. \tag{476}$$

The initial condition requires

$$A_{ik}(0) = P_{ik}, \tag{477}$$

where the P_{ik} are the Schrödinger operators. We now construct

$$\langle \tilde{A} B A \rangle = \left\langle \left(\int^{t} \tilde{\Gamma}(\tau)\, \tilde{K}(t, \tau)\, \mathrm{d}\tau + \tilde{A}_n \right) B \left(\int^{t} K(t, \tau')\, \Gamma(\tau')\, \mathrm{d}\tau' + A_n \right) \right\rangle, \tag{478}$$

where $\sim$ denotes the adjoint matrix. It is assumed that B is a constant matrix

$$B = (B_{kl}) \tag{479}$$

having only one nonvanishing element $(i, j) = (i_1, i_2; j_1, j_2)$:

$$B_{k_1, k_2; l_1, l_2} = \delta_{k_1, i_1} \delta_{k_2, i_2} \delta_{l_1, j_1} \delta_{l_2, j_2}. \tag{480}$$

Using the property (468) we find for the left-hand side of eqn (478) by differentiation

$$\delta_{i_2 j_1} \frac{\mathrm{d}}{\mathrm{d}t} \langle P_{i_1 j_2} \rangle = \delta_{i_2 j_1} \sum_{m,n} \langle M_{i_1, j_2; m, n} P_{m,n} \rangle. \tag{481}$$

By differentiation of the right-hand side of eqn (478) and using the properties of (475) we obtain the following terms

$$\langle \tilde{\Gamma}(t) B A \rangle_{i,j}, \tag{482}$$

$$\langle \tilde{A} B \Gamma(\tau) \rangle_{i,j}, \tag{483}$$

$$\langle \tilde{A} \tilde{M} B A \rangle_{i,j}, \tag{484}$$

$$\langle \tilde{A}BMA\rangle_{i,j}. \tag{485}$$

For the evaluation of expressions (482), (483) we use the Markoffian property, i.e.

$$\langle \Gamma_{i_1,i_2}(t)\,\Gamma_{j_1,j_2}(t')\rangle = G_{i_1,\,i_2,\,j_1,\,j_2}\,\delta(t-t') \tag{486}$$

and

$$\langle \Gamma_{i_1,i_2}(t)\rangle = 0. \tag{487}$$

We now have to perform the average over the heatbath where we know that the fluctuations have only a very short memory. We assume that the response of the particle (or spin) is slow enough so that K and A_n can be taken out of the average (Note that M contains, at most, the heatbath coordinates indirectly over the quantum mechanical ("coherent") fields, but in all loss terms and phase-memory-destroying-parts they do not appear). Equation (482) then gives the contribution

$$G_{i,\,j}/2 \tag{488}$$

and the same quantity similarly follows from (483). In order to evaluate (484) we contract the product of the A's by means of the rule (468) where we make use of the commutativity of the operators Γ_{ik} with the operators of the driving fields within M, which holds in the whole Heisenberg picture. We thus obtain

$$\left\langle \sum_{m,n} P_{mn}\,\tilde{M}_{m,n,i_1,i_2}\,P_{j_1 j_2} \right\rangle = \sum_{m} \langle P_{mj_2}\,M_{i_1,i_2;\,m_1 j_1}\rangle \tag{489}$$

from eqn (484) and a similar one from eqn (485). We now compare the results of both sides, thus obtaining the following equation for the G's which occur in (482), (483)

$$G_{i_1,i_2;j_1,j_2} = \sum_{m,n} \langle \{\delta_{i_2,j_1}\,M_{i_1,j_2;\,m,n} - \delta_{n,j_2}\,M_{i_1 i_2;\,m j_1} - \delta_{m i_1}\,M_{j_1,j_2;\,i_2,n}\}\rangle \langle P_{mn}\rangle. \tag{490}$$

This equation represents the required results. It allows us to calculate all correlation functions of the type (486) if the coefficients M are given, and the solutions P_{ik} of the *averaged* equations are known. A closer inspection† shows that all terms containing external fields cancel each other completely on the right-hand side of eqn (490), so that only the transition rates and damping constants need to be used for the M's. In the following we therefore drop the field terms.

In the case of the laser, the incoherent part of the atomic equations often reads:

† W. Weidlich, private communication.

$$\frac{\mathrm{d}}{\mathrm{d}t}(a_j^\dagger a_j) = \sum_k W_{kj}\, a_k^\dagger a_k - \sum_k W_{jk}\, a_j^\dagger a_j + \Gamma_{jj}(t). \tag{491}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}(a_j^\dagger a_k) = -\gamma_{jk}(a_j^\dagger a_k) + \Gamma_{jk}(t) \qquad (j \neq k). \tag{492}$$

By specializing formula (490) we obtain

$$G_{ii,\,jj} = \delta_{ij}\left\{\sum_k W_{ki}\langle a_k^\dagger a_k\rangle + \sum_k W_{ik}\langle a_i^\dagger a_i\rangle\right\} - W_{ij}\langle a_i^\dagger a_i\rangle - W_{ji}\langle a_j^\dagger a_j\rangle, \tag{493}$$

$$G_{ij,\,ij} = 0, \quad i \neq j \tag{494}$$

$$G_{ij,\,ji} = \sum_k W_{ki}\langle a_k^\dagger a_k\rangle - \sum_k W_{ik}\langle a_i^\dagger a_i\rangle + (\gamma_{ij}+\gamma_{ji})\langle a_i^\dagger a_i\rangle, \quad i \neq j. \tag{495}$$

For many applications (e.g. laser theory) a knowledge of the second moments of the fluctuating forces is completely sufficient, because the fields interact with many independent atoms. Thus the results (e.g. on the linewidth) depend only on a sum over very many independent fluctuating forces which possess a Gaussian distribution (the fluctuating forces of a single atom are not Gaussian, however).†

5.3 THE GENERAL FORM OF THE NON-MARKOFFIAN DENSITY MATRIX EQUATION

We now consider the most general form of a density matrix. We assume that the system considered is subject to external forces, for instance, external electric and magnetic fields, and that it is coupled to external reservoirs (heat-baths). Furthermore we admit that the system itself has an internal interaction, for example, in the case of spins, the spin–spin interaction or in the case of electrons, the electron–electron interaction. These interactions cause transitions between the different quantum states. We assume, as usual, that the equation for ρ is linear in ρ. Then the equation for ρ must be of the following form

$$\rho(t) = \rho(t_0) + \int_{t_0}^{t} K^0(t,\tau)\,\rho(\tau)\,\mathrm{d}\tau + \int_{t_0}^{t} \mathrm{d}\tau \left\{ \sum_{iki'k'} K_{ik,k'i'}(t,\tau)\, P_{ik}\,\rho(\tau)\, P_{k'i'} \right.$$

$$\left. + \sum_{ik} K_{ik}^{(1)}(t,\tau)\, P_{ik}\,\rho(\tau) + \sum_{ik} K_{ik}^{(2)}(t,\tau)\,\rho(\tau)\, p_{ik} \right\} \tag{496}$$

where t_0 is the initial time. As a consequence of causality the integral only runs

† J. R. Senitzky, *Phys. Rev.* **161**, 165 (1967).

from t_0 to t. The most general form in which the projection operators can occur is given by (496) because higher powers of the projection operator can always be reduced according to (466). Provided the kernels K depend only on τ (i.e. $K = K(\tau)$), eqn (496) reduces to an ordinary density matrix equation, which has been introduced, for example, in the theory of magnetic resonance[73] or in the laser.[74] This equation is

$$\frac{\mathrm{d}\rho}{\mathrm{d}t} = \sum_{iki'k'} K_{ik,k'i'}(t)\, P_{ik}\, \rho(t)\, P_{k'i'} + \sum_{ik} K_{ik}^{(1)}(t)\, P_{ik}\, \rho(t) + \sum_{ik} K_{ik}^{(2)}(t)\, \rho(t)\, P_{ik} + K^{(0)}(t)\, \rho(t). \tag{497}$$

5.4. EXACT GENERALIZED FOKKER–PLANCK EQUATION[75]

We define the distribution function by†

$$f(v) = N \int \exp\Big(-\sum_{ik} v_{ik}\, x_{ik}\Big)\, \mathrm{tr}\Big(\prod_{ik} \exp(x_{ik}\, P_{ik})\, \rho\Big)\{\mathrm{d}x_{ik}\}, \tag{498}$$

where N is the normalization constant, and x_{ik} are classical complex quantities for which

$$x_{ik} = -x_{ki}^{*}.$$

The v_{ik}'s are classical variables which are associated with each projection operator:

$$v_{ik} \leftrightarrow P_{ik}; \qquad v_{ik} = v_{ki}^{*}.$$

Most important for what follows is the sequence of the exponential operators which occur in the trace. We order these operators in the following way

$$O = O_L\, O_M\, O_R \equiv \prod_{ik} \exp(x_{ik}\, P_{ik}), \tag{499}$$

where

$$O_L = (1,2)(1,3)(1,4)\ldots(1,n)(2,3)(2,4)\ldots(2,n)\ldots \ldots(n-2,n-1)(n-2,n)(n-1,n) \tag{500}$$

$$O_M = (1,1)(2,2)\ldots(n,n), \tag{501}$$

$$O_R = (n,n-1)(n,n-2)(n-1,n-2)\ldots(n,2)(n-1,2)\ldots \ldots(3,2)(n,1)\ldots(2,1). \tag{502}$$

† This type of distribution function has been introduced by Haken *et al.*[64] where instead of P_{ik}, the usual creation and annihilation operators of Fermions $a_i^{\dagger}$ and a_k were used. In the one particle subspace $a_i^{\dagger}\, a_k$ just obeys the relation (498). The definition (498) secures that f is a real function (which is not always positive, however). (498) allows for a number of modifications and extensions.

We have used the following definitions

$$i < k, \qquad x_{ik} = \xi_{ik}$$
$$(i,k) = \exp(\xi_{ik} P_{ik}) = (1 + \xi_{ik} P_{ik}) \tag{503}$$

$$i = k, \qquad x_{ii} = \xi_{ii} = \eta_i$$
$$(i,k) = \exp(\xi_{ii} P_{ii}) \equiv \exp(\eta_i P_{ii}) \tag{504}$$

$$i > k, \qquad x_{ik} = \tilde{\xi}_{ik}$$
$$(i,k) = \exp(\tilde{\xi}_{ik} P_{ik}) = (1 + \tilde{\xi}_{ik} P_{ik}). \tag{505}$$

The definition (498) allows us to calculate quantum mechanical expectation values by ordinary c-number procedures. For instance the expectation value of P_{lm} is given by

$$\mathrm{tr}\,(P_{lm}\,\rho) = \int \ldots \int \mathrm{d}^N v v_{lm}\, f(v), \tag{506}$$

where the integration runs over all the variables $v_{ik}(= v_{ki}^*)$ over the whole complex plane.

If $\rho(t)$ obeys the usual density matrix equation (497) (which is of first order in its time derivative, the heat baths being Markoffian) it is even possible to calculate multitime averages of the form

$$\mathrm{tr}\,(P_{i_1 k_1}(t_1) P_{i_2 k_2}(t_2) \ldots P_{i_n k_n}(t_n)\, \rho(t_0)), \tag{507}$$

with

$$t_1 > t_2 > \ldots > t_n,$$

by means of the single time distribution function.

The equation for the distribution function

THEOREM: The exact generalized Fokker–Planck equation belonging to (496) and (498) is given by

$$f(t) = f(t_0) + \int_{t_0}^{t} K^{(0)}(t,\tau) f(\tau)\,\mathrm{d}\tau$$
$$+ \int_{t_0}^{t} \mathrm{d}\tau \Big\{ \sum_{ik,i'k'} K_{ik,k'i'}(t,\tau) \sum_{\lambda\mu} M_{k'i',ik,\lambda\mu}(x)\, v_{\lambda\mu}$$
$$+ \sum_{ik} K_{ik}^{(1)}(t,\tau) \sum_{\lambda\mu} N^{(1)}_{ik,\lambda\mu}(x)\, v_{\lambda\mu} + \sum_{ik} K_{ik}^{(2)}(t,\tau) \sum_{\lambda\mu} N^{(2)}_{ik,\lambda\mu}(x)\, v_{\lambda\mu} \Big\} f(\tau). \tag{508}$$

This equation is to be supplemented by the constraint $\sum v_{ii} = 1$. Using the rule

$$x_{ik} \to -\partial/\partial v_{ik}, \tag{509}$$

we define the quantities M, N as follows:

$$M_{ik,k'i';\lambda\mu} = \sum_{mnl} A_{ikmn} B_{k'i'nl} \tilde{C}_{ml\lambda\mu} \exp(x_{nn} - x_{ll}), \tag{510}$$

where

$$A_{ikmn} = \delta_{im} O_L^{k\to n} - x_{mi} \delta_{m<i} O_L^{k\to n}, \tag{511}$$

$$B_{ikmn} = \delta_{kn} O_R^{m\leftarrow i} - x_{kn} \delta_{n<k} O_R^{m\leftarrow i}, \tag{512}$$

$\delta_{m<i} = 1$ for $m < i$ and $= 0$ otherwise.

$$\left.\begin{aligned} &O_L^{m\to k} = \delta_{mk} + \sum x_{ml_1} x_{l_1 l_2} \ldots x_{l_j k}. \\ &\text{The summation runs over all ways from } m \text{ to } k \text{ with} \\ &\qquad m < l_1 < l_2 \ldots < k \qquad \text{for } k \geqslant m \\ &\qquad O_L^{m\to k} = 0 \qquad\qquad \text{for } m > k \end{aligned}\right\} \tag{513}$$

$$\left.\begin{aligned} &O_R^{m\leftarrow l} = \delta_{ml} + \sum x_{ml_j} \ldots x_{l_2 l_1} x_{l_1 l}. \\ &\text{The sum runs over all ways from } l \text{ to } m \text{ with} \\ &\qquad m > l_j > l_{j-1} \ldots > l_2 > l_1 > l \text{ for } m \geqslant l \\ &\qquad O_R^{m\leftarrow l} = 0 \text{ for } m < l \end{aligned}\right\} \tag{514}$$

$$\tilde{C}_{ik,lm} = \begin{cases} C_{iklm} = \delta_{il}(\delta_{km} - \delta_{k<m} x_{km}), & i < k, l < m, \\ D_{iilm} = \delta_{il}\delta_{im}, & i = k, l \leqslant m, \\ E_{iklm} = \delta_{km}(\delta_{il} - x_{li}\delta_{i<l}) \exp(-x_{ii} + x_{kk}), & i > k, l > m, \end{cases} \tag{515}$$

$\tilde{C}_{ik,lm} = 0$ otherwise

$$N^{(1)}_{ikmn} = \begin{cases} H_{ikmm} = \delta_{im}(\delta_{km} - x_{km}\delta_{m<k}), & m = n, i \geqslant k \\ H_{ikmn} = \delta_{im}(\delta_{kn} - x_{kn}\delta_{n<k}) & \\ \qquad + x_{mn}\delta_{n<m}\delta_{in}(\delta_{kn} - x_{kn}\delta_{n<k}), & m > n, i \geqslant k \\ \sum_{m'n'} B_{ikm'n'} \exp(x_{m'm'} - x_{n'n'}) \tilde{C}_{m'n'mn}. & i < k, \end{cases} \tag{516}$$

$N^{(1)}_{ikmn} = 0$ for $i \geqslant k, m < n$ (516a)

$$N^{(2)}_{iklm} = \left\{\begin{aligned} &F_{ikll} = \delta_{il}\delta_{kl} - x_{li}\delta_{l<i}\delta_{kl}, && l = m, i \leqslant k \\ &F_{iklm} = \delta_{km}(\delta_{li} - x_{li}\delta_{l<i}) & \\ &\qquad + \delta_{kl} x_{lm}\delta_{l<m}(\delta_{il} - x_{li}\delta_{l<i}), && l < m, i \leqslant k \\ &\sum_{m'n'} A_{ikm'n'} \tilde{C}_{m'n'lm}, && i > k \end{aligned}\right. \tag{517}$$

$N^{(2)}_{ikmn} = 0$ for $i \leqslant k, l > m$.

Because the operator acting on f on the right-hand side of eqn (508) is linear in $v_{\lambda\mu}$, the Fourier transform of f obeys a partial differential equation which is only of first order in its derivatives. Thus the solution of this equation may offer certain advantages over the direct solution of (508).

Derivation of equation (508)

We first multiply the density matrix eqn (508) with O from the left and then we take the trace over the total system. We then have to calculate the following expressions:

$$\text{tr}\,(OP_{ik}\,\rho P_{n'i'}), \tag{518}$$

$$\text{tr}\,(OP_{ik}\,\rho), \tag{519}$$

$$\text{tr}\,(O\rho P_{ik}). \tag{520}$$

Using the cyclic properties of the trace we can rearrange (518), (519) and (520) in the form

$$\text{tr}\,(P_{k'i'}\,OP_{ik}\,\rho), \tag{521}$$

$$\text{tr}\,(OP_{ik}\,\rho), \tag{522}$$

$$\text{tr}\,(P_{ik}\,O\rho), \tag{523}$$

respectively.

LEMMA 1:

$$O_L{}^{-1} = \left(1 - \sum_{i<k} \xi_{ik} P_{ik}\right). \tag{524}$$

Proof:

We denote the inverse of (i, k) by $\overline{(i, k)}$ so that

$$O_L{}^{-1} = \overline{(n-1, n)}\ \overline{(n-2, n)} \ldots \overline{(l+1, l+2)} \ldots$$
$$\ldots \overline{(l, m+2)}\overline{(l, m+1)}|\overline{(l, m)}\overline{(l, m-1)} \ldots \overline{(1, 2)} \tag{525}$$

if $m+1 \leqslant n$.

If $m = n$, we have instead of (525), the equation

$$O_L{}^{-1} = \overline{(n-1, n)}\overline{(n-2, n)} \ldots \overline{(l+1, l+3)}\overline{(l+1, l+2)}|\overline{(l, n)}\overline{(l, n-1)} \ldots \tag{526}$$

We assume that the expression (524) is correct up to the indices (l, m) i.e. up to the bar, so that we may write

$$O_L{}^{-1} = (1 - \xi_{n-1,n} P_{n-1,n} \cdots - \xi_{ik} P_{ik} \cdots$$
$$- \xi_{l,m+1} P_{l,m+1})|(1 - \xi_{lm} P_{lm}) \ldots \tag{527}$$

if $m+1 \leqslant n$.

If $m = n$, we have instead of (527), the equation

$$O_L{}^{-1} = (1 - \xi_{n-1,n} t P_{n-1,n} \dots - \xi_{ik} P_{ik} \dots - \xi_{l+1,l+2} P_{l+1,l+2})|(1 - \xi_{l,n} P_{l,n}) \dots \tag{528}$$

We now investigate the left neighbour of l, m. According to (525) and (526) the left neighbours have the following indices

$$(l, m+1); (l, m+2) \tag{529}$$

or

$$(l+1, l+2), (l+1, l+3). \tag{530}$$

In any case, we have always

$$m+1, \dots n > l \tag{531}$$

so that the following inequalities for an arbitrary pair (i, k) occurring on the left of the bar result

$$\left.\begin{matrix} i \geqslant l \\ k > i \end{matrix}\right\} \rightarrow k > l \tag{532}$$

From the definition of O_L we have further $m > l$. From these inequalities it is evident that k is not equal to l and thus always

$$P_{ik} P_{lm} = 0 \tag{533}$$

This proves our assertion up to $(l, m-1)$.

LEMMA 2:

$$O_R{}^{-1} = \left(1 - \sum_{i>k} \tilde{\xi}_{ik} P_{ik}\right). \tag{534}$$

Proof:

analogous to that of Lemma 1.

Definitions:

we introduce the decomposition:

$$O_L = O_{LL}^{l,m}(l, m)\, O_{LR}^{l,m}, \tag{535}$$

where

$$O_{LL}^{l,m} = (1, 2)(1, 3) \dots (1, n)(2, 3)(2, 4) \dots (2, n) \dots (l, m-1), \tag{536}$$

if $m > l+1$

and

$$O_{LL}^{lm} = (1, 2)(1, 3) \dots (1, n)(2, 3)(2, 4) \dots (2, n) \dots (l-1, n-1)(l-1, n) \tag{537}$$

if $m = l+1$.

$$O_{LR}^{lm} = (l, m+1)(l, m+2) \ldots (n-2, n-1)(n-2, n)(n-1, n) \quad (538)$$

if $m+1 \leqslant n$,

$$= (l+1, l+2)(l+1, l+3) \ldots (n-2, n-1)(n-2, n)(n-1, n) \quad (539)$$

if $m = n$.

In a similar way we define

$$O_R = O_{RL}^{l,m}(l, m)\, O_{RR}^{l,m} \quad (540)$$

$$O_{RL}^{lm} = (n, n-1)(n, n-2)(n-1, n-2)(n, n-3)(n-1, n-3)(n-2, n-3) \ldots$$
$$\ldots (l+1, m) \quad (541)$$

if $l+1 \leqslant n$,

$$O_{RL}^{lm} = (n, n-1)(n, n-2)(n-1, n-2)(n, n-3)(n-1, n-3)$$
$$.\,(n-2, n-3) \ldots (m+3, m+1)(m+2, m+1) \quad (542)$$

if $l = n$.

$$O_{RR}^{lm} = (l-1, m)(l-2, m) \ldots (m+1, m)(n, m-1)(n-1, m-1) \ldots$$
$$\ldots (m, m-1) \ldots (n, 1)(n-1, 1) \ldots (3, 1)(2, 1), \quad (543)$$

if $l-1 > m$.

$$O_{RR}^{lm} = (n, m-1)(n-1, m-1) \ldots (m, m-1)(n, m-2)(n-1, m-2) \ldots$$
$$\ldots (4, 2)(3, 2)(n, 1)(n-1, 1) \ldots (3, 1)(2, 1) \quad (544)$$

if $l = m+1$.

Simple relations for derivatives

Evidently we have for $l < m$

$$\frac{\partial O}{\partial \xi_{lm}} = \frac{\partial O_L}{\partial \xi_{lm}} O_M\, O_R, \quad (545)$$

with

$$\frac{\partial O_L}{\partial \xi_{lm}} = O_{LL}^{lm}\, P_{lm}\, O_{LR}^{lm}; \quad (546)$$

for $l = m$

$$\frac{\partial O}{\partial \xi_{ll}} = O_L \frac{\partial O_M}{\partial \xi_{ll}} O_R, \quad (547)$$

where

$$\frac{\partial O_M}{\partial \xi_{ll}} = P_{ll}\, O_M = O_M\, P_{ll}; \tag{548}$$

for $l > m$

$$\frac{\partial O}{\partial \tilde{\xi}_{lm}} = O_L\, O_M \frac{\partial O_R}{\partial \tilde{\xi}_{lm}}, \tag{549}$$

with

$$\frac{\partial O_R}{\partial \tilde{\xi}_{lm}} = O_{RL}^{lm}\, P_{lm}\, O_{RR}^{lm}. \tag{550}$$

We now come to the task of expressing (521), (522) and (523) by derivatives of O with respect to x_{ik} (or ξ_{ik}). We do this in several steps: *Rules for shifting projection operators and use of derivatives*

LEMMA 3:

$$P_{ik}\, O = \sum_{mn} A_{ikmn}\, O_L\, P_{mn}\, O_M\, O_R, \tag{551}$$

where A_{ikmn} is defined by (511) and (513).

Proof:

the equation follows from the fact that: (1) the P_{lm}'s span the basis of an algebraic ring, so that we can always express a function of the P_{lm}'s by a linear combination of the P_{lm}'s and that (2) O_L, O_M, O_R possess inverse operators. We multiply (551) by ${O_L}^{-1}$ from the left and by ${O_R}^{-1}\, {O_M}^{-1}$ from the right. This leaves us with the eqn

$${O_L}^{-1}\, P_{ik}\, O_L = \sum_{m,n} A_{ikmn}\, P_{mn}. \tag{552}$$

When we take the matrix element with respect to the states ϕ_m and ϕ_n on both sides, we find immediately

$$A_{ikmn} = \left\langle \phi_m \left(P_{ik} - \sum_{\nu<i} \xi_{\nu i}\, P_{\nu k} \right) O_L\, \phi_n \right\rangle, \tag{553}$$

where we have inserted the explicit form of ${O_L}^{-1}$ according to eqn (524). We further introduce the abbreviation

$$O_L^{m \to k} = \langle \phi_m\, O_L\, \phi_k \rangle. \tag{554}$$

Expanding O_L into the powers of P_{ik} and using the property of the projection operators we immediately find the formula (513).

Introducing (554), (513) into (553) we obtain (511)

LEMMA 4:

$$OP_{ik} = \sum_{mn} B_{ikmn}\, O_L\, O_M\, P_{mn}\, O_R \tag{555}$$

with B_{ikmn} given by (512) and (514).

Proof:

analogous to that of Lemma 3.

We now express $O_L P_{mn} O_M O_R$ by a linear combination of derivatives of O with respect to x_{ik}.

LEMMA 5:

For $i < k$

$$O_L P_{ik} = \sum_{l<m} C_{iklm} \frac{\partial}{\partial \xi_{lm}} O_L \tag{556}$$

with C_{iklm} given in (515)

Proof:

Using (546) we write (556) in the form

$$O_L P_{ik} = \sum_{l<m} C_{iklm}\, O_{LL}^{lm}\, P_{lm}\, O_{LR}^{lm} \tag{557}$$

By multiplying (557) with $O_L{}^{-1}$ from the left and from the right we obtain

$$P_{ik}\, O_L{}^{-1} = \sum_{l<m} C_{iklm}\, (O_{LR}^{lm})^{-1}\, P_{lm}\, (O_{LL}^{lm})^{-1} \tag{558}$$

The right-hand side of eqn (558) follows from

$$O_L{}^{-1}\, O_{LL}^{lm} = (O_{LR}^{lm})^{-1}\, \overline{(l, m)}, \tag{559}$$

$$O_{LR}^{lm}\, O_L{}^{-1} = \overline{(l, m)} (O_{LL}^{lm})^{-1}, \tag{560}$$

(compare (535)) and

$$\overline{(l, m)}\, P_{lm}\, \overline{(l, m)} = P_{lm}, \text{ for } l \neq m. \tag{561}$$

For the further evaluation of the operator product on the right-hand side of eqn (558) we note that

$$\begin{aligned} (O_{LR}^{lm})^{-1}\, P_{lm}\, (O_{LL}^{lm})^{-1} &= -\frac{\mathrm{d}}{\mathrm{d}\xi_{lm}} (O_{LR}^{lm})^{-1}\, \overline{(l, m)}\, (O_{LL}^{lm})^{-1} \\ &= -\frac{\mathrm{d}}{\mathrm{d}\xi_{lm}} O_L{}^{-1}. \end{aligned} \tag{562}$$

Due to (524), (562) reduces to

$$(O_{LR}^{lm})^{-1} P_{lm} (O_{LL}^{lm})^{-1} = P_{lm}. \tag{563}$$

Thus eqn (558) takes the simple form

$$P_{ik} O_L^{-1} = \sum_{l<m} C_{iklm} P_{lm}. \tag{564}$$

Taking the expectation value with respect to the states l and m on the left- and right-hand side we find immediately

$$C_{iklm} = \left\langle \phi_l P_{ik} \left(1 - \sum_{\mu<\nu} \xi_{\mu\nu} P_{\mu\nu}\right) \phi_m \right\rangle. \tag{565}$$

$$C_{iklm} = \delta_{il}\delta_{km} - \delta_{il}\xi_{km}\delta_{k<m} \tag{566}$$

LEMMA 6: for $i = k$

$$O_L P_{ii} O_M = \sum_{l\leqslant m} D_{ii,lm} \frac{\partial}{\partial \xi_{lm}} (O_L O_M) \tag{567}$$

with D given by (515).

Proof:

Use of (546), (548) yields

$$O_L P_{ii} O_M = \sum_{l<m} D_{iilm} O_{LL}^{lm} P_{lm} O_{LR}^{lm} O_M + \sum_l D_{iill} O_L P_{ll} O_M. \tag{568}$$

We multiply (568) from the left with O_L^{-1} and from the right with $O_M^{-1} O_L^{-1}$. This leaves us immediately with

$$P_{ii} O_L^{-1} = \sum_{l<m} D_{iilm} P_{lm} + \sum_l D_{iill} P_{ll} O_L^{-1} \tag{569}$$

which, by use of (524), becomes

$$P_{ii} O_L^{-1} = \sum_{l<m} P_{lm}(D_{iilm} - D_{iill}\xi_{lm}) + \sum_l D_{iill} P_{ll} \tag{570}$$

after the indices have been rearranged. We first take the expectation value with respect to the states l, l which yields

$$D_{iill} = \left\langle \phi_l P_{ii} \left(1 - \sum_{\mu<\nu} \xi_{\mu\nu} P_{\mu\nu}\right) \phi_l \right\rangle = \delta_{il} - \delta_{il} \sum_{l<\nu} \xi_{l\nu}\delta_{\nu l} \tag{571}$$

We now investigate $l \neq m$. Taking the matrix element on the left and right hand side with respect to the states l, m, we find

$$\left\langle \phi_l P_{ii} \left(1 - \sum_{\mu<\nu} \xi_{\mu\nu} P_{\mu\nu}\right) \phi_m \right\rangle = (D_{iilm} - D_{iill}\xi_{lm})\delta_{l<m} \tag{572}$$

and by use of (571) we see that

$$D_{illm} = 0. \tag{573}$$

LEMMA 7: $i > k$

$$O_L P_{ik} O_M O_R = \sum_{l>m} E_{iklm} \frac{\partial}{\partial \xi_{lm}} O = \sum_{l>m} E_{iklm} O_L O_M \frac{\partial}{\partial \xi_{lm}} O_R \tag{574}$$

with E_{iklm} defined in (515).

Proof:

Analogous to that of Lemma 5.

A second way of shifting projection operators:

In the foregoing analysis we have first shifted the projection operators from the left or the right to the middle of the product $O_L O_M O_R$. Then in the next step we have shifted the P_{lm}'s to their places which would correspond to their order in the decomposition (535), (540). Under certain circumstances it leads to simpler formulas if we immediately shift the projection operators to their correct places.

LEMMA 8: $i \leqslant k$

$$P_{ik} O_L O_M = \sum_{l<m} F_{iklm} \left(\frac{\partial}{\partial \xi_{lm}} O_L \right) O_M + \sum_l F_{ikll} O_L \frac{\partial O_M}{\partial \xi_{ll}} \tag{575}$$

where F_{iklm} is given by (517).

Proof:

Multiplying this expression from the left with ${O_L}^{-1}$ and from the right with ${O_M}^{-1} {O_L}^{-1}$, we obtain in the same manner as above, the equation

$${O_L}^{-1} P_{ik} = \sum_{l<m} F_{iklm} P_{lm} + \sum_l F_{ikll} P_{ll} {O_L}^{-1}, \tag{576}$$

and using (524) we get

$${O_L}^{-1} P_{ik} = \sum_{l<m} P_{lm} (F_{iklm} - F_{ikll} \xi_{lm} \delta_{l<m}) + \sum_l F_{ikll} P_{ll}. \tag{577}$$

Equation (577) can be rearranged in the form

$${O_L}^{-1} P_{ik} = \sum_{l<m} F_{iklm} P_{lm} + \sum_l F_{ikll} P_{ll} \left(1 - \sum_{\mu<\nu} \xi_{\mu\nu} P_{\mu\nu} \right) \tag{578}$$

where use of (466) has been made. Taking expectation values on both sides of eqn (578) with respect to states l, m, we obtain immediately (517).

LEMMA 9: $(i \geqslant k)$

$$O_M O_R P_{ik} = \sum_{m \geqslant n} H_{ikmn} \frac{\partial (O_M O_R)}{\partial \xi_{mn}}, \tag{579}$$

where H_{ikmn} is defined in (516).

Proof:
Analogous to that of Lemma 8.

Evaluation of (521), (522), (523)

We are now in a position to put our previous results together in order to calculate the expressions (521), (522) and (523).

1. *Evaluation of* (521)

$$P_{ik}\,OP_{k'i'} = \sum_{m,n} A_{ikmn}\,O_L\,P_{mn}\,O_M\,O_R\,P_{k'i'} \tag{580}$$

(compare (551))
Using

$$O_R\,P_{k'i'} = \sum_{m'n'} B_{k'i'm'n'}\,P_{m'n'}\,O_R, \tag{581}$$

we obtain

$$P_{ik}\,OP_{k'i'} = \sum_{mn}\sum_{m'n'} A_{ikmn}\,B_{k'i'm'n'}\,O_L\,P_{mn}\,O_M\,P_{m'n'}\,O_R. \tag{582}$$

Using

$$O_M\,P_{m'n'} = P_{m'n'}\,O_M \exp(\eta_{m'}-\eta_{n'}), \tag{583}$$

we transform (582) into

$$P_{ik}\,OP_{k'i'} = \sum_{mn}\sum_{m'n'} A_{ikmn}\,B_{k'i'm'n'}\,O_L\,P_{mn}\,P_{m'n'}\,O_M \exp(\eta_{m'}-\eta_{n'})\,O_R, \tag{584}$$

which by use of (466) reduces to

$$\sum_{mnn'} A_{ikmn}\,B_{k'i'nn'}\,O_L\,P_{mn'}\,O_M \exp(\eta_n-\eta_{n'})\,O_R. \tag{585}$$

According to (556), (567) and (574) we have, quite generally, the equation

$$O_L\,P_{mn'}\,O_M\,O_R = \sum_{\lambda\mu} \tilde{C}_{mn'\lambda\mu}\frac{\partial}{\partial x_{\lambda\mu}}\,O, \tag{586}$$

where

$$\tilde{C}_{mn'\lambda\mu} = \begin{cases} C_{mn'\lambda\mu} & \text{for} \quad m < n', \lambda < \mu \\ D_{mn'\lambda\mu} & \text{for} \quad m = n', \lambda \leqslant \mu \\ E_{mn',\lambda\mu} & \text{for} \quad m > n', \lambda > \mu. \end{cases} \tag{587}$$

$$= 0 \text{ otherwise.}$$

Inserting (586) into (585) yields

$$P_{ik}\,OP_{k'i'} = \sum_{mnn'\lambda\mu} A_{ikmn}\,B_{k'i'nn'}\,\tilde{C}_{mn'\lambda\mu}\exp(\eta_n-\eta_{n'})\frac{\partial}{\partial x_{\lambda\mu}}O. \tag{588}$$

Thus, we obtain finally

$$P_{ik}\,OP_{k'i'} = \sum_{\lambda\mu} M_{ikk'i'\lambda\mu}(x)\frac{\partial}{\partial x_{\lambda\mu}}O, \tag{589}$$

with

$$M_{ikk'i'\lambda\mu} = \sum_{mnl} A_{ikmn}\,B_{k'i'nl}\,\tilde{C}_{ml\lambda\mu}\exp(\eta_n-\eta_l). \tag{590}$$

The coefficients A, B, $\tilde{C}$, are given by the expressions (511), (512) and (515).

2. *Evaluation of* (523)

We distinguish between the cases $i \leqslant k$ and $i > k$. If $i \leqslant k$ we apply (575) If $i > k$ we first use (551) which yields

$$P_{ik}\,O = \sum_{mn} A_{ikmn}\,O_L\,P_{mn}\,O_M\,O_R. \tag{591}$$

Inserting

$$O_L\,P_{mn}\,O_M\,O_R = \sum_{m'l'} \tilde{C}_{mnm'l'}\frac{\partial}{\partial x_{m'l'}}O \tag{592}$$

into (591) we find

$$P_{ik}\,O = \sum_{lm}\frac{\partial O}{\partial x_{lm}}\mathscr{T}_{lm},$$

where

$$\mathscr{T}_{lm} = \sum_{m'n'} A_{ikm'n'}\,\tilde{C}_{m'n'lm}. \tag{593}$$

3. *Evaluation of* (522)

In a similar way we treat OP_{ik}. For $i > k$ we apply (579). For $i < k$ we have

$$OP_{ik} = O_L\,O_M\sum_{mn} B_{ikmn}\,P_{mn}\,O_R. \tag{594}$$

We find

$$OP_{ik} = \sum_{mn} B_{ikmn}\,O_L\,P_{mn}\exp(\eta_m-\eta_n)\,O_M\,O_R. \tag{595}$$

Inserting (586), that is

$$O_L\,P_{mn}\,O_M\,O_R = \sum_{l'm'} \tilde{C}_{mnl'm'}\frac{\partial}{\partial x_{l'm'}}O, \tag{596}$$

into (594), we obtain

$$OP_{ik} = \sum_{l'm'} N_{ikl'm'} \frac{\partial}{\partial x_{l'm'}} O \tag{597}$$

with

$$N_{ikl'm'} = \sum_{mn} B_{ikmn} \exp(\eta_m - \eta_n) \tilde{C}_{mnl'm'}. \tag{598}$$

Final step

We insert the explicit expressions for (521), (522) and (523) into the equation

$$\operatorname{tr}(O\rho(t)) = \operatorname{tr}(O\rho(t_0)) + \int_{t_0}^{t} d\tau \left\{ \sum_{iki'k'} K_{ikk'i'}(t,\tau) \operatorname{tr}(P_{k'i'} OP_{ik} \rho(\tau)) \right.$$
$$\left. + \sum_{ik} K_{ik}^{(1)}(t,\tau) \operatorname{tr}(OP_{ik} \rho(\tau)) + \sum_{ik} K_{ik}^{(2)}(t,\tau) \operatorname{tr}(P_{ik} O\rho(\tau)) \right\}. \tag{599}$$

In order to obtain an equation for the distribution function f, (498), we multiply (599) by

$$\exp\left\{ -\sum_{ik} v_{ik} x_{ik} \right\}$$

and integrate over the space $\{x_{ik}\}$. Thus the left-hand side of (599) becomes identical with $f(t)$. The traces on the right-hand side of eqn (599) have the form of derivatives of O with respect to x_{ik}, which are multiplied by functions of x_{ik}. These operations can be translated immediately into ones with respect to the variables v_{ik}, if the following wellknown rules are observed:

$$\partial/\partial x_{ik} \rightarrow \text{multiplication by } v_{ik}$$
$$\text{multiplication by } x_{ik} \rightarrow -\partial/\partial v_{ik} \tag{600}$$

Having these rules in mind we immediately find eqn (508) which was presented in 5.4.

In conclusion we discuss a constraint which stems from the normalization of any quantum system, which is expressed by

$$\sum_i P_{il} = 1. \tag{601}$$

We include (601) explicitly in f (498) by the following new definition of f,

$$f = N \int \{\mathrm{d}x_{ik}\} \exp\left\{-\sum_{ik} v_{ik}\, x_{ik}\right\}$$

$$\times \frac{1}{2\pi} \int_{-\infty}^{\infty} \mathrm{d}\xi \exp(-i\xi)\, \mathrm{tr}\left(O_L \exp\left(i\xi \sum_j P_{jj}\right) O_M\, O_R\, \rho\right). \tag{602}$$

Having in mind that $O_M = \exp\{x_{jj} P_{jj}\}$ we introduce the new variables $\tilde{x}_{jj} = x_{jj} + i\xi$ which leaves us with

$$f = N \int \{\mathrm{d}x_{ik}\} \exp\left\{-\sum_{i \neq k} v_{ik}\, x_{ik}\right\} \exp\left\{-\sum_j v_j\, \tilde{x}_{jj}\right\} \mathrm{tr}\,(O\rho)$$

$$\times \frac{1}{2\pi} \int \exp\left(i\xi \left(\sum_j v_{jj} - 1\right)\right) \mathrm{d}\xi. \tag{603}$$

Evidently the definition (498) of f agrees with that of (603) if the constraint $\sum_j v_{jj} = 1$ is added to the definition (498).

Many subsystems: projection on to macroscopic variables

Our eqn (508) applies to a single atom with two energy levels, or a spin$\frac{1}{2}$, to multi-level atoms, as well as to a complicated many-body problem. For a discussion of its solution it is thus certainly necessary to treat specific examples.† On the other hand, under certain circumstances it is possible to perform strong simplifications even in rather general cases. Indeed, in many cases of practical importance, the system under consideration consists of many equivalent subsystems. Examples are spins in external fields, or laser atoms interacting with the light field. In these examples the individual subsystems interact with the external fields in a similar way, i.e. by their total (macroscopic) electric or magnetic moments (after certain phase-factors have been transformed away). In these cases it is advantageous, first to introduce projection operators for each subsystem μ: $P_{ik,\mu}$ and to formulate the density matrix equation by means of these $P_{ik,\mu}$. It reads

$$\dot{\rho}(t) = L\rho(t), \tag{604}$$

where L is a linear operator (see eqn (496)). In our present case, L consists of a sum over the subsystems:

$$L = \sum_{\mu} \tilde{L}_{\mu}(P_{ik\mu}) \equiv \sum_{\mu} \tilde{L}(P_{ik\mu}), \tag{605}$$

where the latter equation expresses the fact that the subsystems are equivalent.

† e.g. in the case of a two-level atom, f is built up of δ-functions (W. Weidlich, private communication).

We now introduce macroscopic variables

$$V_{ik} = \sum_{\mu} v_{ik\mu} \tag{606}$$

and define

$$f(t) = \int \{\mathrm{d}x_{ik}\} \exp\left\{-\sum_{ik} x_{ik} V_{ik}\right\} \mathrm{tr}\left(\left(\prod_{\nu} O_{\nu}\right) \rho\right), \tag{607}$$

where

$$O_{\nu} = \prod_{ik} \exp\left(x_{ik} P_{ik\nu}\right). \tag{608}$$

Because the O_{ν}'s commute, we may write

$$\prod_{\nu} O_{\nu} = \prod_{\nu}^{(\mu)} O_{\nu} \,.\, O_{\mu},$$

where (μ) indicates, that the factor $\nu = \mu$ is to be omitted. We multiply eqn (604) from the left with O and take the trace with respect to the whole system, thus

$$\begin{aligned} \mathrm{tr}\,(O\rho) = \mathrm{tr}\,(OL\rho(\tau)) &= \sum_{\mu} \mathrm{tr}\,(O\tilde{L}(P_{ik\mu})\rho) \\ &= \sum_{\mu} \mathrm{tr}^{(\mu)} \left[\left(\prod_{\nu}^{(\mu)} O_{\nu}\right) \mathrm{tr}_{\mu}\left(O_{\mu}\, \tilde{L}(P_{ik\mu})\rho\right)\right]. \end{aligned} \tag{609}$$

It has been shown that

$$\mathrm{tr}_{\mu}\left(O_{\mu}\, \tilde{L}(P_{ik\mu})\rho\right) = h\left(x_{ik}, \frac{\partial}{\partial x_{ik}}\right) \mathrm{tr}_{\mu}\left(O_{\mu}\, \rho\right). \tag{610}$$

Note that O_{μ} is a function of x_{ik} due to (608). The right-hand side of eqn (609) can thus be written in the form

$$(609) = \sum_{\mu} \mathrm{tr}^{(\mu)} \left[\left(\prod_{\nu}^{(\mu)} O_{\nu}\right) h\left(x_{ik}, \frac{\partial}{\partial x_{ik}}\right) \mathrm{tr}_{\mu}\left(O_{\mu}\, \rho\right)\right] \tag{611}$$

$$= \sum_{\mu} \mathrm{tr}\left[\prod_{\nu}^{(\mu)} O_{\nu} \,.\, h\left(x_{ik}, \frac{\partial}{\partial x_{ik}}\right) O_{\mu}\, \rho\right]. \tag{612}$$

Note that we can always use the cyclic properties of the traces, if needed.

Now let us make use of the fact that h is of first order in the derivatives $\partial/\partial x_{ik}$ (compare eqns (508) and (600)). From Leibniz's rule for differentiating

products, we immediately find

$$\mathrm{tr}\,(O\rho) = h\left(x_{ik}, \frac{\partial}{\partial x_{ik}}\right) \mathrm{tr}\left(\prod_{\nu} O_\nu\, \rho\right). \tag{613}$$

Multiplying eqn (613) with

$$\exp\left(-\sum_{ik} x_{ik}\, V_{ik}\right)$$

and integrating over x_{ik}, we obtain

$$f(V_{ik}, t) = \chi\left(V_{ik}, \frac{\partial}{\partial V_{ik}}\right) f(V_{ik}, \tau), \tag{614}$$

where χ is a linear operator defined on the right-hand side of eqn (508), if the v_{ik}'s are replaced by the V_{ik}'s. A simple analysis shows that the condition $\sum_i v_{ii} = 1$ is now to be replaced by $\sum V_{ii} = N$, where N is the number of subsystems. The importance of the result (614) is that while $f(v_{ik}, t)$ for a single subsystem may show wild fluctuations (e.g. it consists of δ-functions), the macroscopic variables V_{ik} possess a smooth distribution, which changes but little. Thus in many cases of interest it can be shown that we may neglect derivatives higher than second order, so that (614) then actually reduces to an ordinary Fokker–Planck equation.

Appendix

Quantum mechanical Langevin equations[76]

We shall discuss the origin of quantum mechanical Langevin forces for the field (the effect of heatbaths).

The light field

We shall show by means of an example, which can be treated exactly, how quantum mechanical Langevin forces arise. For this end we couple the light-field to a heat-bath which consists of an infinite number of harmonic oscillators. Thus the Hamiltonian of the bath has the form

$$\sum_{\omega} \hbar\omega\, B_\omega^\dagger\, B_\omega. \tag{615}$$

The bath variable occurring in the interaction Hamiltonian (297) is chosen as a superposition

$$B = \hbar \sum g_\omega\, B_\omega, \tag{616}$$

or, in the interaction representation,

$$B(t) = \hbar \sum_{\omega} g_{\omega} B_{\omega} \exp(i\omega t). \tag{617}$$

The total interaction Hamiltonian has the form[75]

$$H_{\text{field bath}} = b^{\dagger} \sum_{\omega} g_{\omega} B_{\omega} \exp(i\omega t) + b \sum_{\omega} g_{\omega}^{*} B_{\omega}^{\dagger} \exp(-i\omega t). \tag{618}$$

The Hamiltonian of the free field is given by

$$\hbar\omega_0 b^{\dagger} b. \tag{619}$$

The Heisenberg equations for $b_{\lambda}^{\dagger}$ and $B_{\lambda}^{\dagger}$ are

$$\mathrm{d}b^{\dagger}/\mathrm{d}t = i\omega_0 b^{\dagger} + i \sum_{\omega} g_{\omega}^{*} B_{\omega}^{\dagger} \exp(i\omega t). \tag{620}$$

and

$$\mathrm{d}B_{\omega}^{\dagger}/\mathrm{d}t = ib^{\dagger} g_{\omega} \exp(-i\omega t). \tag{621}$$

Equation (621) has the solution

$$B_{\omega}^{\dagger} = i \int_{t_0}^{t} b^{\dagger}(\tau)\, g_{\omega} \exp(-i\omega\tau)\, \mathrm{d}\tau + B_{\omega}^{\dagger}(t_0), \tag{622}$$

where $B_{\omega}^{\dagger}(t_0)$ is the operator at time t_0. Inserting (622) into (620) we arrive at

$$\frac{\mathrm{d}b^{\dagger}}{\mathrm{d}t} = i\omega_0 b^{\dagger} - \int_{t_0}^{t} b^{\dagger}(\tau) \sum_{\omega} |g_{\omega}|^2 \exp[i\omega(t-\tau)]\, \mathrm{d}\tau + i \sum_{\omega} g_{\omega}^{*} B_{\omega}^{\dagger}(t_0) \exp(i\omega t). \tag{623}$$

We now assume that the g_{ω}'s are of about equal amplitudes so that we have

$$\int_0^{\infty} |g|^2 \exp[i\omega(t-\tau)]\, \mathrm{d}\omega = \kappa \left[\delta(t-\tau) + \frac{i}{\pi} \frac{P}{t-\tau} \right] \tag{624}$$

$$\kappa = \pi |g|^2$$

If we allow also for negative frequencies, then for harmonic oscillators with an inverted energy scale we would find, instead of (624), the equation

$$\int_{-\infty}^{+\infty} |g|^2 \exp[i\omega(t-\tau)]\, \mathrm{d}\omega = 2\kappa\, \delta(t-\tau). \tag{625}$$

For the evaluation of the integrals it must be noted that the δ-function has the property

$$\int_0^t \delta(t-\tau)\, \mathrm{d}\tau = \tfrac{1}{2}. \tag{626}$$

Equation (624) corresponds to the choice made by Senitzky, whereas the choice (625) was made by Lax. We see that (624) is more physical but (625)

has certain computational advantages. The actual difference between (625) and (624) is negligible in practical calculations. Under these assumptions (623) becomes

$$\frac{db^\dagger}{dt} = i\omega_0 b^\dagger - \kappa b^\dagger + \underbrace{i\sum_\omega g_\omega^* B_\omega^\dagger(t_0)\exp(i\omega t)}_{F^\dagger(t)}, \tag{627}$$

where the last term is evidently the fluctuating force.

We determine the properties of this fluctuating force and evaluate

$$\begin{aligned}\langle F^\dagger(t)F(t')\rangle_B &= \sum_\omega |g_\omega|^2 \exp[i\omega(t-t')]\,\mathrm{tr}\,(B_\omega^\dagger B_\omega \exp[-H_B^{(\omega)}/k\tau])Z_\omega^{-1}\\ &= \sum_\omega |g_\omega|^2 \exp[-i\omega(t-t')]\,\bar{n}_\omega(T)\end{aligned} \tag{628}$$

and correspondingly

$$\langle F(t)F^\dagger(t')\rangle_B = \sum_\omega |g_\omega|^2 \exp[-i\omega(t-t')]\,(\bar{n}_\omega(T)+1), \tag{629}$$

where $\bar{n}_\omega$ is the occupation number of the mode at the frequency ω of the heatbath. The bath average over the commutator has the form

$$\langle [F(t)F^\dagger(t')]\rangle_B = \begin{cases}\kappa\left(\delta(t-t') + \dfrac{i}{\pi}\dfrac{P}{t'-t}\right)\\ 2\kappa\delta(t-t')\end{cases} \tag{630}$$

Equations (628) and (629) can be directly expressed by the damping constant κ only for the temperature $T = 0$. For higher temperatures it must be noted that in practical calculations

$$\langle F^\dagger(t)F(t')\rangle = \sum_\omega |g_\omega|^2 \exp[i\omega(t-t')]\,\langle B_\omega^\dagger B_\omega\rangle, \tag{631}$$

and

$$\langle F(t)F^\dagger(t')\rangle = \sum_\omega |g_\omega|^2 \exp[i\omega(t-t')]\,\langle B_\omega B_\omega^\dagger\rangle, \tag{632}$$

always appear under an integral which contains a factor $\exp(i\omega_0 t)$, where ω_0 is the frequency of the harmonic oscillator (light field). If this is taken into account, (628) may be case into the form

$$\sum_\omega |g_\omega|^2 \exp[i\omega(t-t')]\,\bar{n}_\omega(T) = 2\kappa\,\bar{n}_{\omega_0}(T)\,\delta(t-t'), \tag{633}$$

and similarly

$$\langle F(t)F^\dagger(t')\rangle = 2\kappa(\bar{n}_{\omega_0}(T)+1)\,\delta(t-t'). \tag{634}$$

The δ-function in (630) and (633) expresses the fact that the heatbath has a very short memory. As may be seen from (623) and (624) it is essential for the derivation that the heatbath frequencies have a spread which covers a whole range around the oscillator frequency ω_0. We solve eqn (620) explicitly

$$b^\dagger = b^\dagger(0) \exp(i\omega_0 - \kappa)\, t + \int_0^t \exp[(i\omega_0 - \kappa)(t-\tau)]\, F^\dagger(\tau)\, \mathrm{d}\tau \tag{635}$$

and get

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle b^\dagger b\rangle_B = \langle \dot{b}^\dagger b + b^\dagger \dot{b}\rangle_B$$

$$= -2\kappa\langle b^\dagger b\rangle_B + \int_0^t \langle F^\dagger(t)\, F(\tau)\rangle_B \exp[-(i\omega_0 + \kappa)(t-\tau)]\, \mathrm{d}\tau$$

$$+ \int_0^t \langle F^\dagger(\tau)\, F(t)\rangle_B \exp(i\omega_0 - \kappa)(t-\tau)\, \mathrm{d}\tau. \tag{636}$$

The integrals may be evaluated using (633) and (634) so that we find

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle b^\dagger b\rangle_B = -2\kappa\langle b^\dagger b\rangle_B + 2\kappa\bar{n}. \tag{637}$$

In analogy to (636) we form

$$\frac{\mathrm{d}}{\mathrm{d}t}\langle [b, b^\dagger]\rangle_B = -2\kappa\langle [b, b^\dagger]\rangle + 2\kappa. \tag{638}$$

The general solution reads

$$\langle [b, b^\dagger]\rangle_{B,t} = \exp(-2\kappa t)\, C + 1. \tag{639}$$

If we insert the initial condition

$$\langle [b, b^\dagger]\rangle_{B,0} = 1, \tag{640}$$

we find

$$C = 0, \tag{641}$$

which means that the commutation relation is preserved for all times.

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Acknowledgements

I wish to extend my sincere thanks to all who have helped me to bring the manuscripts into their final form. I mention in particular Mrs. U. Funke, Miss S. M. Kay and Mr. A. Maitland. I further owe thanks to Drs. Graham, Reineker, Sauermann and Vollner for their help in reading the proofs.

Optical Resonators

G. Toraldo di Francia

1. Introduction

The threshold value of the pump power in a typical laser is given by

$$W_{\text{pump}} \geqslant \frac{phv}{\phi t_{\text{phot}}} \tag{1}$$

where p represents the number of modes which can interact with the emission line of the material, ϕ is the quantum efficiency, and t_{phot} denotes the time during which a photon can interact with the material. W_{pump} can be reduced by making p small and t_{phot} large. Such favourable conditions can be achieved by using a resonant cavity having a high value of Q ($= 2\pi\nu t_{\text{phot}}$) and chosen so that only a small number of modes (possibly only one) interacts with the atomic systems. Resonant cavities have been widely employed in microwave devices and these techniques have now been applied to optical systems.

A closed cavity with perfectly conducting walls has an infinite number of resonant modes of different frequencies. If we want only one mode to resonate within a fairly wide frequency band, the size of the cavity must be of the same order of magnitude as the wavelength, and it is practically impossible to achieve this at optical frequencies. When the size of the cavity is much larger than the wavelength, the number of modes per unit volume which can resonate in a frequency interval $d\nu$ is $(8\pi\nu^2/c^3)\,d\nu$. For example, a 1 cm^3 cavity used at optical frequencies would have more than 10^8 modes, even within a bandwidth, $d\nu/\nu$, of 10^{-6}.

However, a closed cavity with perfectly conducting walls represents a very idealised situation. In order to use the field inside the cavity, it must have an opening through which energy can be transmitted to the rest of the system. If the losses are high, the value of Q is reduced and the width of the resonance curve increases. If Q is small for a whole set of modes whose frequencies are closely spaced, then the separate frequencies will merge into a continuum. Strictly speaking, a lossy cavity does not have discrete modes, but only pseudo-modes or virtual modes. The situation is analogous to the case, in quantum mechanics, of a system of particles with positive energy. Roughly speaking,

the meaning of a pseudo-mode is that when a photon is in such a state, it has a high probability of staying inside the cavity for a long time. In the case of an ordinary microwave cavity with waveguide terminals, however, it is still fairly accurate to speak of discrete modes.

The situation is different for an open resonator, i.e. for a cavity where a substantial part of the walls has been removed. A large majority of the modes have very low Q and merge into a continuum. However, it was pointed out by Schawlow and Townes[1] that if the size of the cavity is very large compared with the wavelength, as necessarily happens in optics, a few particular modes may have relatively low losses, due to the directivity of the radiation. The triple infinity of modes which can resonate in a closed cavity reduce to a single infinity, and it may then be possible to have only one or a few modes resonating within the atomic linewidth.

The active material filling a laser cavity may substantially influence the modes, but for most cases, we will ignore this influence and consider the resonator to be situated in free space.

2. The Fabry–Pérot Cavity

The above reasoning was first applied to a Fabry–Pérot cavity, Here the radiation bounces back and forth between two parallel flat mirrors. One or both end mirrors are made slightly transparent, to provide a coupling with external space.

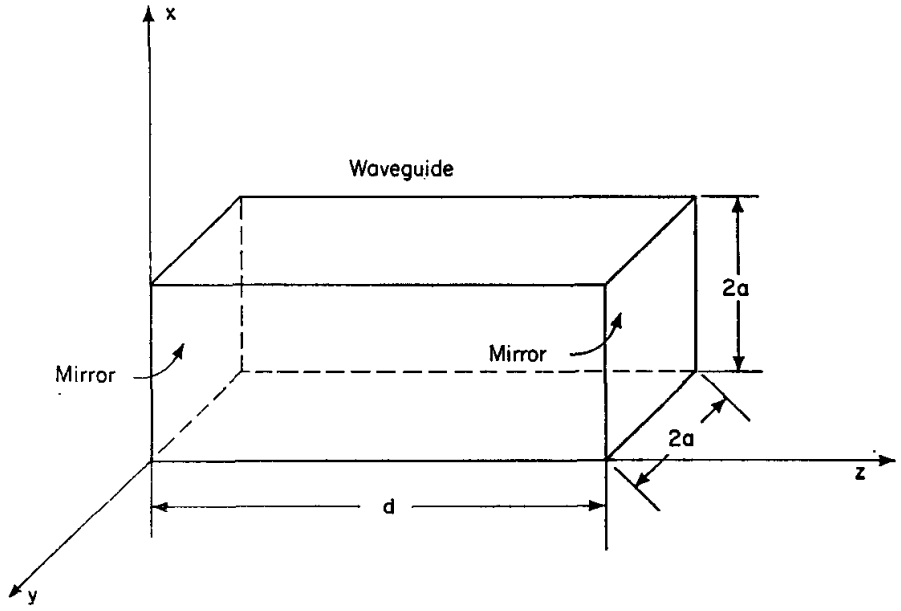


FIG. 1. Diagram showing the two mirrors of a Fabry–Pérot cavity forming the end bases of a waveguide, of length d and square cross-section of dimension $2a$.

An approximate theory of this multimode resonator was proposed by Schawlow and Townes.[1] They assumed that the two mirrors form the end bases of a section of a waveguide with metal walls (Fig. 1). Let the waveguide have length d (along the z-axis) and a square cross-section of dimension $2a$ (in the xy-plane). The frequencies of the modes of oscillation of this rectangular cavity have the following form

$$\nu = \frac{c}{2n}\left[\left(\frac{q}{d}\right)^2 + \left(\frac{r}{2a}\right)^2 + \left(\frac{s}{2a}\right)^2\right]^{\frac{1}{2}}, \tag{2}$$

where q, r, s are positive integers, c is the velocity of light and n is the refractive index of the medium. Each mode can be generated by a plane wave reflected back and forth between the walls [2] and characterized by the direction cosines $\pm r\lambda/4a$, $\pm s\lambda/4a$, $\pm q\lambda/2d$, where λ is the wavelength in the medium.

For the present purpose, we need consider only those waves which make a very small angle with the axis of the waveguide and which radiate almost end-on through the slightly transparent mirrors. For such waves, q is approximately equal to the number of half wavelengths contained in d, and has a very large value (10^5–10^6), while r and s are small. In this case, if the ratio $2a/d$ is not too small, (2) can be written as

$$\nu = \frac{c}{2n}\left(\frac{q}{d} + \frac{r^2 d}{8qa^2} + \frac{s^2 d}{8qa^2}\right). \tag{3}$$

If we denote the reflection coefficient of the mirrors by α, a nearly-axial wave will decay in a time t given by

$$t = \frac{nd}{(1-\alpha)c}. \tag{4}$$

Hence

$$Q = \frac{2\pi d}{(1-\alpha)\lambda}. \tag{5}$$

If $\alpha = 0{\cdot}98$ and $d = 1$ cm, Q is of the order of 10^7.

Now we have from (3) that two modes with values of q differing by 1 will be separated in frequency by approximately

$$\Delta\nu_q = \frac{c}{2nd}. \tag{6}$$

If α is close to unity, it follows from (4) that $\Delta\nu_q \gg 1/t$. This means that successive axial modes are discrete. If the bandwidth of the transition is less than $c/2nd$, it will be possible to set a single axial mode into oscillation. Alterna-

tively, discrimination against unwanted axial modes can be achieved by an extra reflecting surface suitably positioned outside the laser.[3]

However, modes with the same value of q and successive values of r are separated in frequency by an amount,

$$\Delta\nu_r = c\frac{(r+\frac{1}{2})d}{8nqa^2}\,, \tag{7}$$

and a corresponding result may be found for s. A typical value for $d/2a$ in a solid state laser is 10. Hence, with $\alpha = 0{\cdot}98$ and r of the order of unity, we have $\Delta\nu_r < 1/t$. This means that the lateral modes are almost degenerate and may merge into a continuum. They may therefore be set into laser oscillation simultaneously.

In principle, lateral modes may be separated by their directional properties. Each mode will radiate into four directions, characterised by the angles $\pm r\lambda/4a$, $\pm s\lambda/4a$. Hence the directions of radiation of the modes form a regular array of spacing $\lambda/4a$. However, the finite size of the end apertures will give rise to diffraction, each beam having a half-width given approximately by $\lambda/4a$. It would therefore be just possible to isolate a single mode by focussing the radiation onto a screen with a suitable pinhole.

Let us now remove the side walls and deal with a true Fabry–Pérot resonator. For small values of r and s, the modes are very nearly the same as those of the closed cavity, the only difference is that, due to the absence of the side walls, they continually spill energy into external space. This will reduce the Q of all modes, and the effect will be more pronounced, the higher the values of r and s. The resonator may loose energy in two ways; firstly, by reflection, oblique waves after being reflected back and forth will gradually walk off the end mirrors, and secondly, by diffraction.

The time t' for a beam to walk off the end mirror is readily found to be given by

$$t' = \frac{8na^2}{rc\lambda}\,. \tag{8}$$

This is equal to the decay time t, given by (4) when

$$r = \frac{8a^2(1-\alpha)}{\lambda d}\,. \tag{9}$$

For $\alpha = 0{\cdot}98$, $d = 10$ cm, $2a = 0{\cdot}5$ cm, $\lambda = 5 \times 10^{-5}$ cm, we obtain $r = 20$. Hence, modes with $r > 20$ have a Q that is less than half the Q of the fundamental mode. If the pump power is not too high, these will be below threshold and will not oscillate. However, we can conclude, even before taking diffrac-

tion into account, that a few tens or hundreds of lateral modes are likely to lase at the same time.

The first satisfactory treatment of the Fabry–Pérot resonator was given by Fox and Li.[4] They treated the waves as scalar quantities. This approach can be justified since the size of the mirrors is large compared with the wavelength of the light and all angles of interest are sufficiently small.

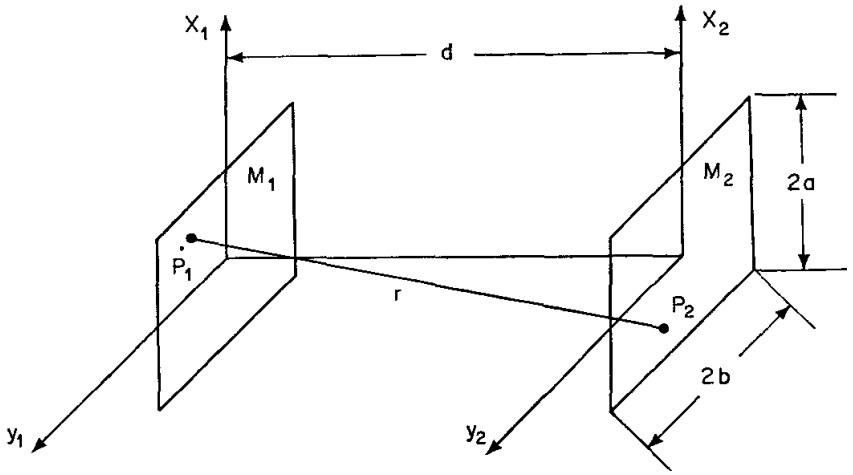


FIG. 2. Diagram showing the arrangement of mirrors, M_1 and M_2, of a Fabry–Pérot resonator.

Their treatment can be summarised as follows. Let us assume that the two mirrors M_1 and M_2 (Fig. 2) are identical rectangles. If we start with a field distribution $u_1(x_1, y_1)$ across M_1, by Huygens' principle we will obtain a distribution $u_2(x_2, y_2)$ across M_2, given by

$$u_2(x_2, y_2) = \frac{i}{\lambda}\int_{M_1} u_1(x_1, y_1)\frac{\exp(ikr)}{r}\cos\theta\, ds_1 \tag{10}$$

where θ is the angle between P_1P_2 and the normal to the mirrors, r denotes the distance from (x_1, y_1) to (x_2, y_2), $k = 2\pi/\lambda$ and a factor $\exp(-i\omega t)$ has been omitted.

The normal modes or eigenfunctions of the resonator are obtained by requiring that the field distribution over M_1 reproduces itself over M_2, so that $u_2 = \sigma u_1$, where σ is a constant factor. As a result, we obtain the integral equation

$$\sigma u(x_2, y_2) = \frac{i}{\lambda}\int_{M_1} u(x_1, y_1)\frac{\exp(ikr)}{r}\cos\theta\, dx_1\, dy_1. \tag{11}$$

Each eigenfunction u represents a mode of the resonator and the eigenvalue σ specifies both the attenuation and the phase change in one trip across the resonator.

When the mirror separation d is much larger than the lateral dimensions a, b, eqn (11) can take the simpler form

$$\sigma u(x_2, y_2) = \frac{i}{\mathrm{d}\lambda} \int_{M_1} u(x_1, y_1) \exp\left[ik \frac{(x_1-x_2)^2+(y_1-y_2)^2}{2d}\right] \mathrm{d}x_1\, \mathrm{d}y_1 \quad (12)$$

where a factor exp (ikd) corresponding to the normal optical path has been omitted on the right hand side. The argument of the complex number σ will now be called the *phase shift*.

Equation (12) is valid when $a^2/d\lambda \ll d^2/a^2$, where a represents a typical dimension of the mirrors. If we introduce the *Fresnel number*

$$N = \frac{a^2}{\mathrm{d}\lambda}, \quad (13)$$

we can write this condition as†

$$N \ll \frac{d^2}{a^2}. \quad (14)$$

By introducing the dimensionless variables $\xi_{1,2} = \sqrt{N}\,\frac{x_{1,2}}{a}$, $\eta_{1,2} = \sqrt{N}\,\frac{y_{1,2}}{a}$, eqn (12) becomes

$$\sigma u(\xi_2, \eta_2) = i \int_{\mathscr{M}_1} u(\xi_1, \eta_1) \exp\{i\pi[(\xi_1-\xi_2)^2+(\eta_1-\eta_2)^2]\} \mathrm{d}\xi_1\, \mathrm{d}\eta_1 \quad (15)$$

where $\mathscr{M}_1$ represents the domain of ξ_1, η_1 space corresponding to the domain M_1 of x_1, y_1 space. It can be seen by inspection that for a set of geometrically similar mirrors, eqn (15) depends only on N (through $\mathscr{M}_1$).

In the case of the rectangular mirrors of Fig. 2, we can separate the variables, by putting $u(\xi, \eta) = u(\xi)\, v(\eta)$. The integral equation for u becomes

$$\sigma u(\xi_2) = \exp(-i\pi/4) \int_{-\sqrt{N}}^{+\sqrt{N}} u(\xi_1) \exp[i\pi(\xi_1-\xi_2)^2]\, \mathrm{d}\xi_1. \quad (16)$$

It is obvious that u and v represent solutions for infinite-strip mirrors, parallel to y and x respectively. Thus, the eigenfunctions and eigenvalues for the rectangular mirror are products of the infinite-strip eigenfunctions and eigenvalues respectively. Specifying the different eigenfunctions by subscripts, we shall write $u_{mn}(\xi, \eta) = u_m(\xi)\, v_n(\eta)$ and $\sigma_{mn} = \sigma_m \sigma_n$.

Equation (16) is a homogeneous Fredholm equation of the second kind with $u(\xi)$ as the kernel. The eigenfunctions are orthogonal over the surface of the mirror.

† Actually a closer investigation shows that condition (14) is not as strictly necessary as might appear. Equation (12) turns out to be valid under much more general conditions.[5-7]

Fox and Li[4] have solved the integral equation numerically, by an iteration method. Starting from an arbitrary initial field at M_1, one can compute the field produced over M_2 by Huygens' principle. This new field distribution is then used to compute the field produced at M_1 as a result of the second transit. By repeating this calculation over and over again, one finds that the relative field distribution approaches a steady state. Depending on whether the initial field is symmetric or antisymmetric, one ends up with the lowest symmetric or antisymmetric mode.

Figure 3 shows the results obtained by Fox and Li[4] for the lowest even (symmetric) and odd (antisymmetric) modes, for different values of N. The power loss and phase shift are both found to be very rapidly decreasing functions of N. At resonance, the separation d of the mirrors is an integral number of half-wavelengths plus a very small additional amount corresponding to the phase shift. The numerical analysis has confirmed that the wave guide approach is a fairly good approximation and can help to understand several qualitative features.

A very elegant approach to the theory of the Fabry–Pérot resonator was given by Wainstein.[8] The flat mirrors are treated as the walls of a two-di-

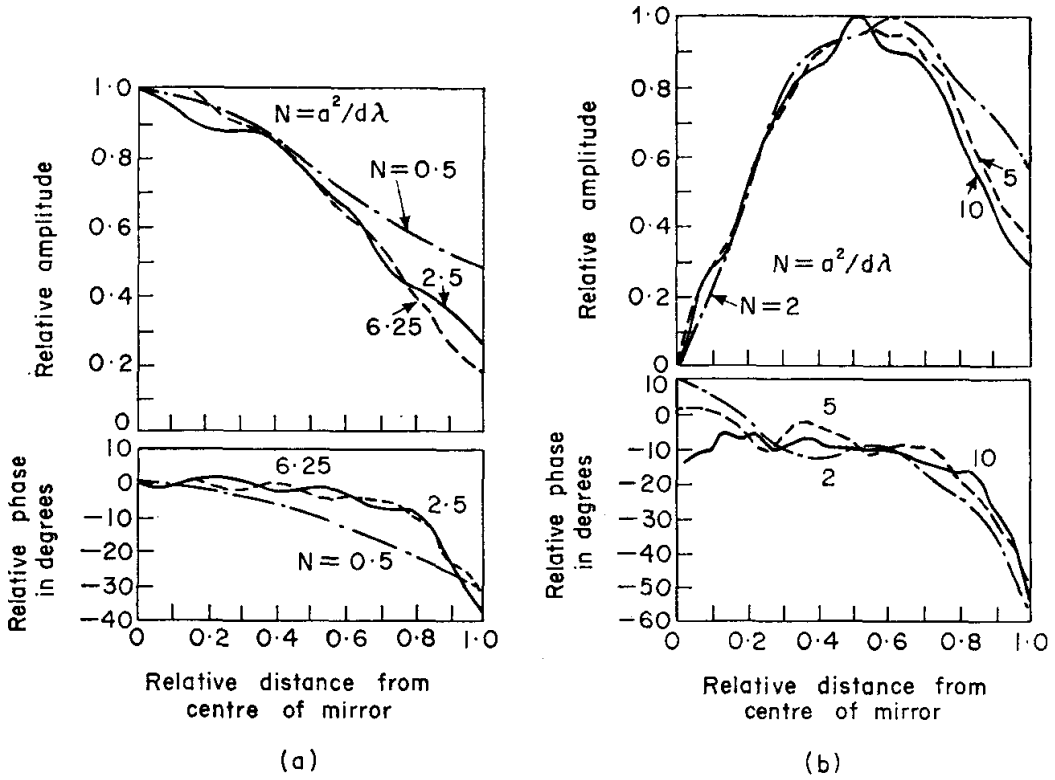


FIG. 3. Curves showing the amplitudes and phases of the field distributions using different values of N, for:

(a) the lowest even (symmetric) mode;
(b) the lowest odd (antisymmetric) mode.

(After Fox and Li[4].)

mensional waveguide, open at both ends (Fig. 4). A travelling mode very near cut-off consists of a plane wave travelling almost perpendicular to the walls and reflected back and forth by the walls. When the wave arrives at an open end, it is almost entirely reflected back into the same mode travelling in the opposite direction. In this way the condition for axial resonance of the Fabry–Pérot resonator becomes a condition for lateral resonance. By applying the theory of open-end waveguides, Wainstein was able to derive approximate expressions for the field distributions, the losses and resonant frequencies. The overall agreement with the results of Fox and Li is very good. The merit of Wainstein's theory is that one can obtain expressions for the quantities of interest in closed form.

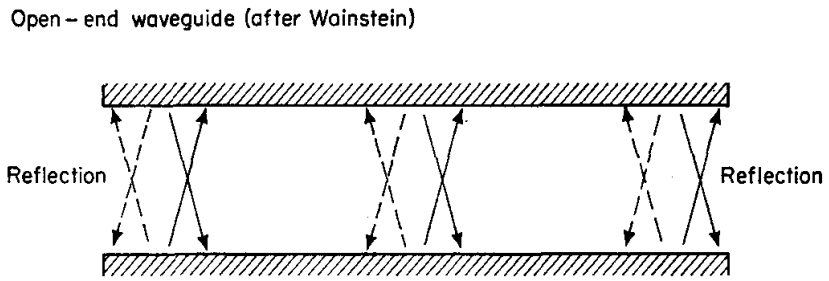


FIG. 4. Following Wainstein's approach, the mirrors are treated as the walls of a 2-*d* waveguide.

It is of practical interest to consider the case where the medium between the mirrors is non-linear (saturable gain). This case can be dealt with by assuming the non-linear medium to be concentrated in two infinitely thin slabs close to the mirrors. Equation (11) can be suitably modified to solve this problem[6–11] and it turns out that the presence of the non-linear medium alters the modal phase distributions, without appreciably affecting the amplitude distributions.

The iterative method is especially suitable for the determination of the lowest-loss even and odd modes. Determination of higher order modes, while not impossible, is rather cumbersome, and to make such a determination easier, Fox and Li[12] and Wells[13] have elaborated other numerical techniques. The principle of the Fox and Li method, called "resonance excitation" is as follows. A resonator is illuminated with a fixed frequency through one of the end mirrors and the length d is varied until resonance is reached for a prescribed mode. The field distribution across the opposite mirror turns out to contain predominantly that mode. If a number of identical resonators are arranged in series and suitably separated by isolators, the field distribution across the output mirror virtually coincides with the prescribed mode. In practice, a sequence of three resonators is sufficient for determining a mode.

Alternative approaches to the theory of the Fabry–Pérot resonator are based on the use of variational techniques either in coordinate or momentum space.[14-18] These approaches must be applied with some caution, otherwise they may lead to inaccurate results. When correctly applied, they become very laborious and require the solution of a system of many homogeneous linear equations and the associated determinantal equation. Risken,[19] however, produced an accurate solution quite simply.

Another interesting approach is to consider a mode of an open resonator as a free oscillation of the system.[20-23] One looks for the conditions (a complex frequency) under which an outgoing field can exist in the absence of an incoming field. This approach can give results in quite good agreement with those found by Fox and Li.

Some computations have been made for the case when the plane end mirrors are not perfectly parallel, but are slightly tilted.[24, 16] It turns out that the power losses of the modes increase very rapidly with the tilt angle, especially for large values of N. Even a very small tilt, corresponding to departure from parallelism of a fraction of a wavelength at the edge, can give rise to substantial loss. This means that the alignment of the plane-parallel resonator is very critical. Moreover, the lowest order modes tend to become degenerate in loss. They will be present at the same time in the laser and will give rise to beats at the detector.

Experimental verification of the behaviour of a Fabry–Pérot resonator is very difficult to carry out at optical frequencies, mainly because of the tremendous accuracy with which the resonator must be built and adjusted in order to obtain significant results. However, a very accurate experiment can be carried out with microwaves.[25, 26] If the mirrors are flat to within $\lambda/300$, the mode distribution coincides with that described by Fox and Li.

3. The Confocal Resonator

A typical confocal resonator is formed by two spherical convex mirrors with a common focus. There is, however, a large family of systems which behave optically in the same way as the confocal resonator.[27, 28] We will first outline the general properties of this family. Let two plane mirrors M_1, M_2 (Fig. 5) be placed at the focal planes, Σ_1, Σ_2 of an ideal (free from aberrations) optical system S. The light beams in both directions will be assumed to be limited solely by the finite sizes of the mirrors.

A given field distribution $u_1(x_1, y_1)$ across Σ_1 will give rise by inverse interference to a set of plane waves, each one specified by the first two direction cosines α, β of its propagation vector. The complex amplitude of a wave at the

origin O_1 will be denoted by $A(\alpha, \beta)\, d\alpha\, d\beta$ and can be evaluated by the Fourier transform[29]

$$A(\alpha, \beta) = \frac{1}{\lambda^2} \int_{M_1} u_1(x_1, y_1) \exp\left[-ik(\alpha x_1 + \beta y_1)\right] dx_1\, dy_1. \tag{17}$$

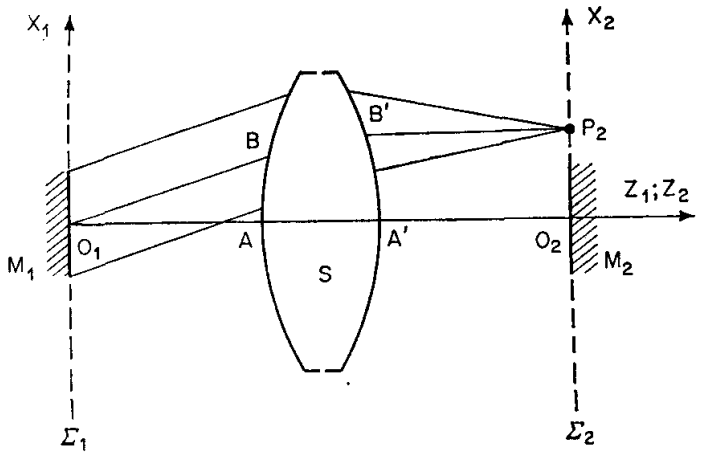


FIG. 5. Representation of a confocal resonator system. Σ_1 and Σ_2 are the focal planes of the optical system S.

System S will focus each plane wave $A(\alpha, \beta)$ onto a point P_2 of Σ_2. Since S has been assumed to be perfect, it will obey the sine condition.[30] Thus the coordinates of P_2 are

$$x_2 = f\alpha, \qquad y_2 = f\beta, \tag{18}$$

where f represents the focal length of S.

It can be shown [30a] that the energy carried by the wave is equal to $C\lambda^2|A|^2\, d\alpha\, d\beta$, where C is a constant of proportionality. If u_2 is the amplitude of the field across Σ_2, then the energy carried by the wave is equal to $C|u_2|^2 dx_2\, dy_2$. From eqn (18) the area illuminated, $dx_2\, dy_2$, is equal to $f^2\, d\alpha\, d\beta$. Hence $|u_2| = \lambda|A|/f$. To find the phase, we note that the optical path $O_1BB'P_2$ of any wave $A(\alpha, \beta)$ is identical to the optical path $O_1AA'O_2$ of the axial wave; this optical path will be denoted by d. Moreover, we must take into account the effect of anomalous propagation, [30b] by which a spherical wave arrives at its focus with a phase shift of $-\pi/2$. Hence we can write

$$u_2(x_2, y_2) = -i\lambda \frac{\exp(ikd)}{f} A(\alpha, \beta). \tag{19}$$

If M_1 and M_2 are equal and we require that u_2 be proportional to u_1, then we

find the following integral equation for the field distribution u on any one of the mirrors;

$$\sigma u(x_2, y_2) = \frac{\exp i(kd - \pi/2)}{\lambda f} \int_{M_1} u(x_1, y_1) \exp\left[-\frac{ik}{f}(x_1 x_2 + y_1 y_2)\right] dx_1 dy_1 \tag{20}$$

We will assume the contour of the mirrors to be symmetrical about the x and y axes. It is then an easy matter to prove that the eigenfunctions of (20) can then be split into a set of modes symmetrical about the origin and a set of modes antisymmetrical about the origin. The integral (20) turns out to be real for a symmetric mode and pure imaginary for an antisymmetric mode. Hence we find that the total phase shift for a round trip in the interferometer is $2kd - \pi$ for a symmetric mode and $2kd - \pi \pm \pi$ for an antisymmetric mode. Consequently, symmetric modes will resonate for

$$d = q\frac{\lambda}{2} + \frac{\lambda}{4} \tag{21}$$

and antisymmetric modes for

$$d = q\frac{\lambda}{2}, \tag{22}$$

where q is an integer.

The modes turn out to be highly degenerate, and can only be discriminantly selected if they have different diffraction losses.

Solutions of eqn (20), in particular cases, have been discussed by Goubau and Schwering[31] and by Boyd and Gordon.[32]

It is convenient to include the factor $\exp[-i(kd - \pi/2)]$ in σ, to introduce the Fresnel number $N = a^2/f\lambda$ and to introduce reduced coordinates analogous to those of the previous section. Equation (20) can then be written in standard form

$$\sigma u(\xi_2, \eta_2) = \int_{M_1} u(\xi_1, \eta_1) \exp[-2\pi i(\xi_1 \xi_2 + \eta_1 \eta_2)] \, d\xi_1 d\eta_1. \tag{23}$$

In the case of rectangular mirrors the variables can be separated and eqn (23) splits into two Fredholm equations of the second kind with symmetric kernels, each equation being valid for an infinite strip mirror.

The equation for the ξ coordinate becomes

$$\sigma_m u_m(\xi_2) = \int_{-\sqrt{N}}^{+\sqrt{N}} u_m(\xi_1) \exp(-2\pi i \xi_1 \xi_2) \, d\xi_1. \tag{24}$$

For obvious reasons, the u_m are called the eigenfunctions of the finite Fourier transform. They turn out to be angular prolate spheroidal functions.[33] The use of these functions in optical problems has been extensively discussed in recent years.[34–43]

When the mirrors are circular, one can introduce polar coordinates and in place of (24) one finds a finite Hankel transform. The eigenfunctions are then hyperspheroidal functions.[37, 44]

In both cases it is found that, at least for the first eigenmodes, the "spot size" (i.e. the size of the region illuminated on the end mirror) is very small compared with the size of the mirror. The intensity falls off exponentially from the centre and is virtually negligible at the rim. This explains why the losses are extremely small, orders of magnitude smaller than in the Fabry–Pérot case.

Since the finite size of the mirror has an almost negligible influence on the mode configuration, it can be disregarded in a simplified theory. One then finds Hermite–Gaussian eigenfunctions for the infinite strip case[32] and Laguerre–Gaussian eigenfunctions for the circular case.[31] This will be discussed later.

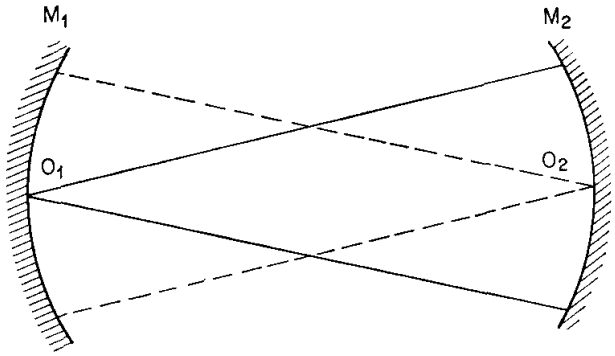


FIG. 6. Confocal system of mirrors.

We find first of all that the system S can be replaced by two lenses, each one very close to one of the mirrors; then each system, mirror-plus-lens, can be replaced by a spherical mirror as shown in Fig. 6. Each mirror has its centre of curvature on the other mirror and the system is confocal.

4. Beam Waveguides and Non-Confocal Resonators

Let us start with a set of cylindrical coordinates r, ϕ, z (Fig. 7) and transform these to spheroidal coordinates ξ, ϕ, θ, using the following relations

$$r = c\sqrt{(1+\xi^2)}\sin\theta, \qquad \phi = \phi, \qquad z = c\xi\cos\theta, \tag{25}$$

where c represents a constant length.

The surfaces $\xi = \textit{constant}$ represent a set of confocal oblate spheroids, while the surfaces $\theta = \textit{constant}$ represent a set of confocal single-sheet hyperboloids.

The geometrical foci FF' (to be distinguished from the "optical" foci) are separated by a distance $2c$. The major and minor semi-axes of each ellipse are given by $a = c(+\xi^2)^{\frac{1}{2}}$, $b = c\xi$; from which the radius of curvature of the ellipse at the intersection with the z axis is found to be $R = a^2/b = c(1+\xi^2)/\xi$. The major and minor semi-axes of each hyperbola are given by $a = c \sin\theta$, $b = c \cos\theta$, where θ is the angle between the asymptotes and the z axis.

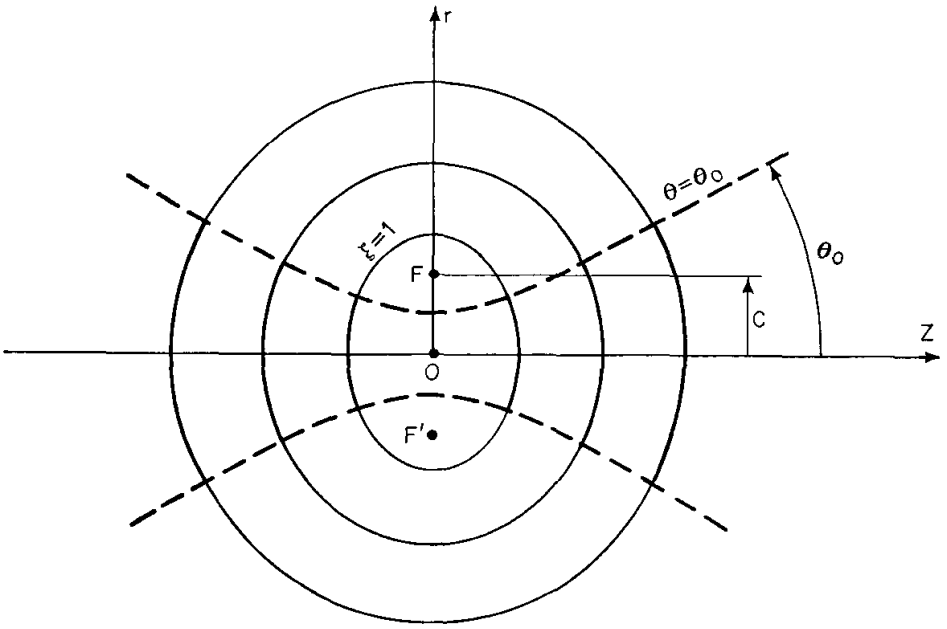


FIG. 7. Transformation of cylindrical coordinates to spheroidal coordinates. Surfaces of constant ξ represent a set of confocal oblate spheroids and surfaces of constant θ represent a set of confocal hyperboloids.

We can write the wave equation in spheroidal coordinates (25). Wainstein showed[45] that it can be solved approximately when the following conditions are satisfied;

$$kc \gg 1, \qquad \sin^2\theta \ll 1. \tag{26}$$

These conditions hold if:

(1) the distance FF' is much greater than the wavelength and

(2) propagation is confined within a small solid angle about the axis.

In real systems these conditions are usually very well satisfied.

We introduce a new variable

$$\tau = \sqrt{(2kc)}\,\theta. \tag{27}$$

A solution of the wave equation with separated variables can then be written

in the form

$$u_n^m(\xi, \tau, \phi) = (1+\xi^2)^{-\frac{1}{2}} \exp\{i[kc\xi - (2n+m+1)\arctan\xi]\} \times \cos(m\phi)\, \tau^m L_n^m\left(\frac{\tau^2}{2}\right) \exp\left(-\frac{\tau^2}{4}\right), \quad (28)$$

where L_n^m denotes an associated Laguerre polynomial. This expression clearly represents a travelling wave, whose wave surfaces coincide with the spheroids $\xi = constant$.

The last factor of eqn (28) shows that the field decreases exponentially away from the axis. We can define a "spot size" s, which represents a region about the axis where the intensity of the radiation is non-negligible.

$$s = \left[\frac{c(1+\xi^2)}{k}\right]^{\frac{1}{2}}. \quad (29)$$

From this definition it is seen that propagation is virtually confined within a single-sheet hyperboloid. Although this hyperboloid represents an ideal case, it does show that the system acts like a waveguide. For this reason, it is customary to speak of a "beam waveguide". A wave described by eqn (28) will be called a "beam" for short.

If one of the spheroidal surfaces of Fig. 7 is replaced by the surface of a mirror, the beam will be reflected at this surface and will exactly retrace its steps, taking on the same configuration at all positions. If two spheroidal surfaces are replaced by mirrors, we obtain a resonator. Because the spot size is so small it is permissible for the mirrors

(1) to be spherical instead of spheroidal, and

(2) to have a finite size.

We thus have a general resonator with spherical mirrors.

The confocal system is formed by the two mirrors $\xi = 1$. In the language of geometrical optics, the hyperboloid of Fig. 7 would be called a "caustic" surface. The existence of caustic surfaces has been proved for resonators of different geometries,[46-51] for example, toroidal-ring resonators. The existence or non-existence of a caustic surface can be directly related to the stability or instability of the mode.

The condition for resonance is found from (28) by requiring that the phase shift in going from the first to the second mirror should be a multiple of π. All modes having the same value of $2n+m$ have the same resonance (degeneracy).

The cartesian coordinate x can be used in eqn (25) in place of the coordinate r. A solution of the wave equation, independent of y, can then be found. This is the solution for infinite strip mirrors, which turns out to be a Hermite–

Gaussian expression instead of a Laguerre–Gaussian expression, as was mentioned in the previous section.

Products of two infinite strip modes (one for x and one for y) can be used in place of the Laguerre–Gaussian modes. As long as the mirrors are very large in comparison with s and there is no perturbation present within the resonator, these two sets of modes are perfectly equivalent (degenerate). The finite size of the mirrors can act as a perturbation which removes this degeneracy. If the mirrors are circular, the modes are Laguerre–Gaussian functions, while for rectangular mirrors the modes are Hermite–Gaussian functions. In a conventional gas laser the main perturbation is usually due to the Brewster angle windows. This leads to separation of the x and y variables i.e. to Hermite–Gaussian modes. Mode patterns of the type shown in Fig. 8 are obtained.

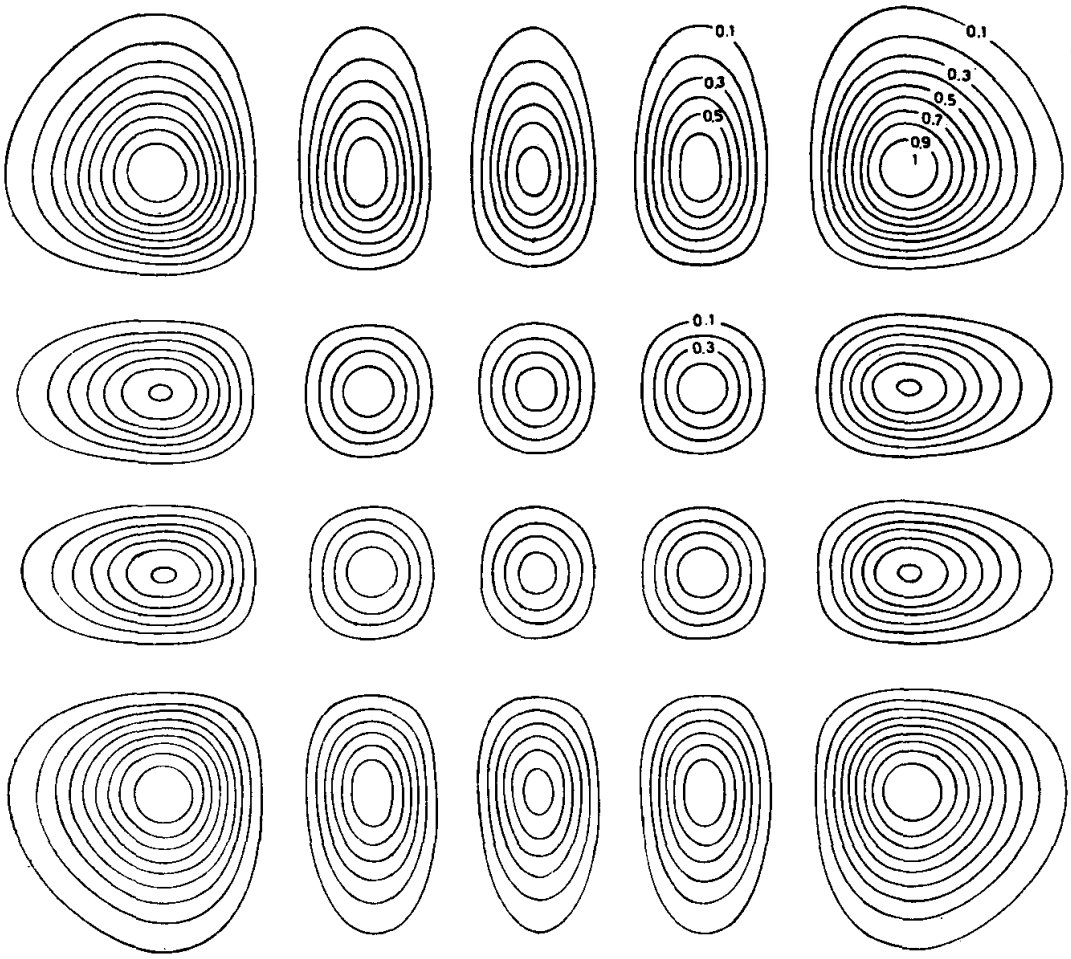


FIG. 8. Example of a mode pattern in a gas laser with Brewster angle windows.

Equation (28) describes the field inside a resonator when the diffraction due to the finite size of the mirrors is neglected. The equation may also be used to calculate the losses, but it has been proved by Wainstein [52, 53] that this leads to an over estimation of the losses. Hence we conclude that diffraction at the edges of the mirrors alters the modal field distributions in such a way as to reduce the losses.

It is clear that, apart from the dependence on ϕ, a beam is completely specified by the location of its centre O (Fig. 7) and of one of its geometrical foci, say F. Any other element can be derived by a graphical construction, suggested by Deschamps and Mast.[54] Let the beam be specified by the centre O and the focus F (Fig. 9) separated by a distance c. We want to find the relevant parameters of the wave surface through the point M on the axis.

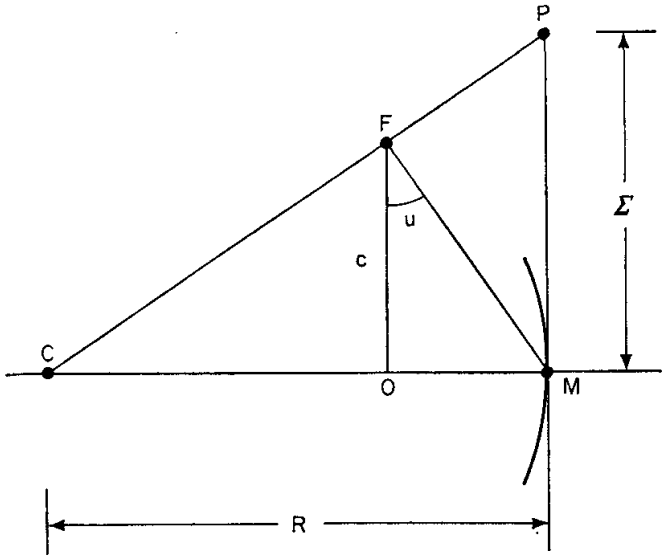


FIG. 9. Graphical construction suggested by Deschamps and Mast for finding the parameters of a beam.

If $OM = c\xi$, the wave surface has the equation $\xi = OM/c$. Let us put

$$\tan u = \xi, \qquad R = \frac{c}{\sin u \cos u}, \qquad \Sigma = \frac{c}{\cos^2 u}. \tag{30}$$

Therefore $u = M\hat{F}O$. Using the value of u we can immediately calculate the phase shift in expression (28):

$$\text{phase shift} = (2n+m+1)\,u. \tag{31}$$

The quantity R is easily found to be equal to $c(1+\xi^2)/\xi$, i.e. to the radius of curvature of the wave surface at M. It is seen by inspection that the centre of curvature C of the wave surface can be found by drawing the perpendicular to

MF through F. As for the quantity Σ, it turns out that its value $c(1+\xi^2)$ is proportional to the square of the spot size. Conventionally, Σ will be called the "beam area". The beam area is found by inspection to be equal to MP.

By this graphical construction or by other types of equivalent transformations[55-57] one can easily solve a number of practical problems.

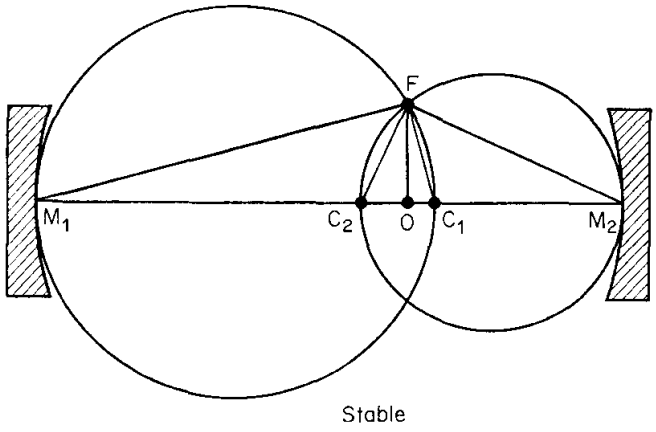


FIG. 10. Arrangement of a resonator with spherical mirrors corresponding to a stable configuration.

First of all, let us determine the parameters of the beam which travels back and forth within a resonator with spherical mirrors. Let C_1 and C_2 (Fig. 10) represent the centres of curvature of the mirrors. They must coincide with the centres of curvature of the wave surfaces of the beam at M_1 and M_2 respectively. Obviously, one has to draw the two circles shown in the figure; their intersection F determines the geometrical focus of the beam and O its centre. When a point of intersection can be found in this way, the configuration is "stable". An "unstable" configuration is shown in Fig. 11. All pairs of mirrors which give rise to the same beam are "equivalent".

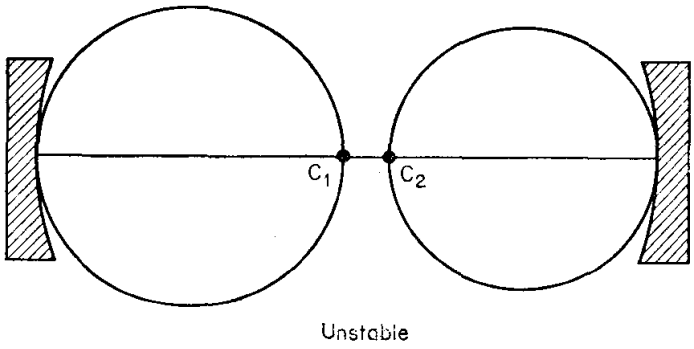


FIG. 11. Arrangement corresponding to an unstable configuration

In the confocal configuration, shown in Fig. 12, the two circles coincide and any point on the circumference can be labelled as F. However, it is clear that the symmetrical location of F, shown in the figure, is the one which gives minimum beam area on the mirrors. This situation corresponds to minimum loss, and takes place in practice.

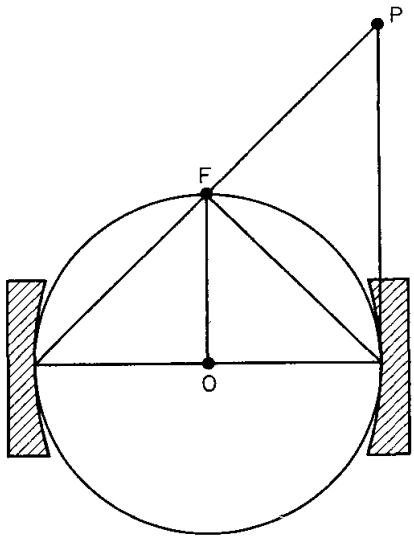


FIG. 12. Configuration in a confocal resonator corresponding to minimum loss.

The graphical construction is useful for describing the passage of a beam through an optical system, say a thin lens (Fig. 13). Let the incident beam be specified by its characteristic points O and F. We want to find the characteristic points O', F' of the image beam. First, we find the centre of curvature C of the wave incident upon the lens. Obviously, the image of C is at C' according to the usual thin lens formula. Next, we find the point P which determines the beam area on the plane of the lens; P' must coincide with P. It is now an easy matter to complete the construction and find F' and O'.

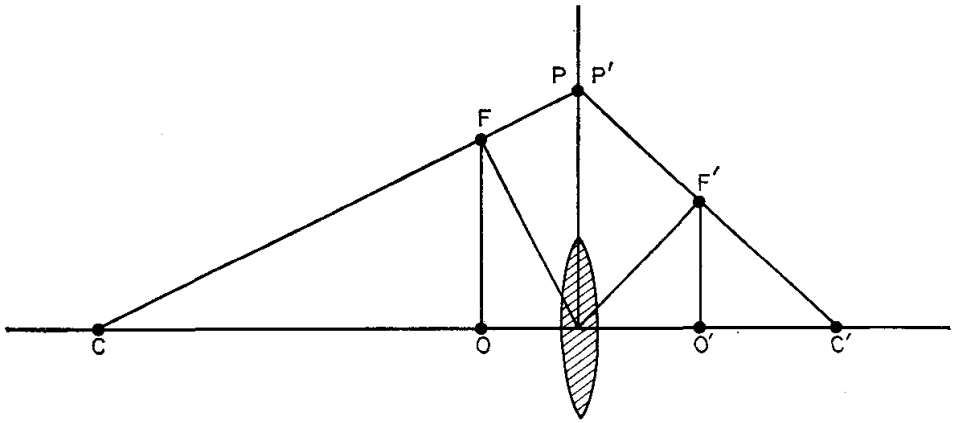


FIG. 13. Graphical construction for describing the passage of a beam through a thin lens.

5. Periodic Systems and Stability

Since the early days of open resonators, it has been noted that the theory of an open resonator is in many respects equivalent to that of a travelling-wave system in a periodic structure.

For instance, we can start with the resonator of Fig. 5 and "unfold" it, by substituting real spaces for the virtual spaces created by the mirrors M_1, M_2. We thus arrive at the travelling-wave system depicted in Fig. 14, where the mirrors are replaced by periodic aperture stops M_0, M_1, M_2... .

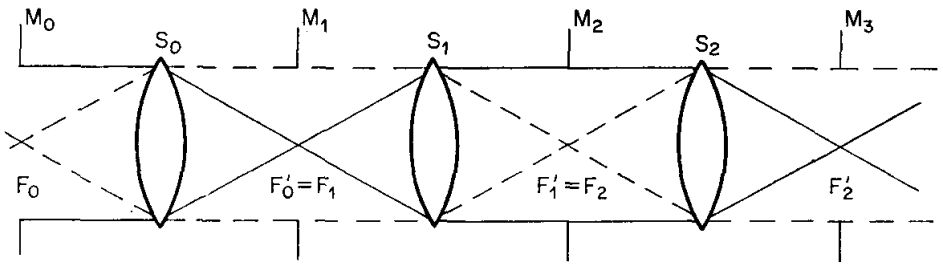


Fig. 14. Travelling-wave system equivalent to a confocal resonator.

Each pair of consecutive lenses, say S_0, S_1, is equivalent to an afocal system of unit magnification. The rear focus F_0' of S_0 coincides with the front focus F_1 of S_1. The separation d between the rear principal plane of S_0 and the front principal plane of S_1 is equal to twice the focus length. In a chain of such systems each ray repeats its path periodically with respect to the lenses.

If the spacing d is now changed from twice the focal length, the pair of lenses S_0, S_1 is no longer afocal. However, it may be possible for a set of n consecutive subsystems S_0, S_1, ..., S_{n-1} to form an afocal system of unit magnification. The following analysis, due to Burch[58] shows that this is possible.

Let the spacing of the identical systems S_n, whose focal length will be taken as unity, be such that F'_{n-1} lies in front of F_n by a distance α. We take an object point located on the axis at a distance x_0 to the left of the front focus F_0 of S_0. The image point will be located at a distance $x_0' = 1/x_0$ to the right of F_0' and consequently at a distance $x_1 = \alpha - 1/x_0$ to the left of F_1. We thus obtain the recurrent relation

$$x_1 = \alpha - \frac{1}{x_0}, \qquad x_2 = \alpha - \frac{1}{x_1}, \qquad \ldots . \tag{32}$$

After traversing n subsystems, the displacement x_n is given by the continued fraction

$$\frac{1}{x_n} = \cfrac{1}{\alpha - \cfrac{1}{\alpha - \cfrac{1}{\alpha - \cfrac{1}{\ddots \cfrac{}{\alpha - \cfrac{1}{x_0}}}}}}, \tag{33}$$

where α appears n times.

For the system to be afocal, we require $x_0 = x_n = \infty$ and so obtain an equation for α of degree $(n-1)$, whose solutions can be shown to be given by

$$\alpha = 2\cos\left(\frac{r}{n}\pi\right), \tag{34}$$

where $r = 1, 2, \ldots, n-1$. There are therefore $n-1$ distinct solutions. However, it will be noted that among the solutions for n there are also solutions for the submultiples of n. The lateral magnification, which equals $(-1)^n/(x_0 x_1 \ldots x_{n-1})$ turns out to be $(-1)^r$.

If we now assign a value to α, we must distinguish between two cases. If $|\alpha| > 2$, there are no values of r and n which satisfy eqn (34). Indeed, as is well known in optical design and in accelerator theory, if the spacing between the foci exceeds twice the focal length, the system is unstable[59,60] and any ray traversing the system gradually moves away from the axis and will eventually fall outside the system. If $|\alpha| < 2$, it may be possible to find a pair of integers r, n which satisfy eqn (34) and then we have an n-fold afocal system. However, even if such a pair of numbers does not exist, the fraction r/n can be chosen as close as desired to any real number. We can conclude that any physical system with $|\alpha| < 2$ is practically n-fold afocal, provided that n is made sufficiently large.

Let us now consider an open resonator with spherical mirrors M_1, M_2, having radii of curvature R_1, R_2 (positive for concave mirrors, negative for convex mirrors) and spacing d. This system is equivalent to a succession of thin lenses having alternatively a focal length $f_1 = R_1/2$, and $f_2 = R_2/2$. A subsystem S_n will be formed by a pair of successive lenses. Since the focal length of the subsystem will be taken to be unity we find

$$\frac{1}{f_1} + \frac{1}{f_2} - \frac{d}{f_1 f_2} = 1 \tag{35}$$

The distances l, l' from the front focus to the first lens and from the second lens to the rear focus, are given by

$$l = 1 - \frac{d}{f_2}, \qquad l' = 1 - \frac{d}{f_1}. \tag{36}$$

We will now introduce two convenient parameters

$$g_1 = 1 - \frac{d}{R_1}, \qquad g_2 = 1 - \frac{d}{R_2}, \tag{37}$$

or

$$g_1 = 1 - \frac{d}{2f_1}, \qquad g_2 = 1 - \frac{d}{2f_2}. \tag{38}$$

From these definitions and from (35) we find

$$d = 2g_1 + 2g_2 - 4g_1 g_2. \tag{39}$$

The distance α $(= d - l - l')$ from the rear focus of one subsystem to the front focus of the next one becomes

$$\alpha = 2 - 4g_1 g_2, \tag{40}$$

For an n-fold system we have therefore from (34)

$$g_1 g_2 = \sin^2\left(\frac{r}{n}\frac{\pi}{2}\right). \tag{41}$$

For stability, the product $g_1\, g_2$ must be positive and less than 1. We can plot a stability diagram (Fig. 15) as was done by Boyd and Kogelnik,[60] and by Fox and Li.[24]

The coordinate axes represent g_1 and g_2. The shaded regions correspond to high loss or unstable systems, while the unshaded regions, lying between the axes and the hyperbola $g_1\, g_2 = 1$ represent low loss or stable systems. Points representing mirrors with equal radii lie on the 45° line $g_1 = g_2$. The origin corresponds to identical mirrors spaced confocally, the point (1, 1) corresponds to plane parallel mirrors and the point $(-1, -1)$ corresponds to concentric mirrors. Confocal systems lie on the dashed hyperbola $2g_1\, g_2 - g_1 - g_2 = 0$ and it is seen that they are all unstable, except for the system with identical mirrors. However, it must be emphasised that the confocal resonator with identical mirrors lies on the boundary between stable and unstable regions. Consequently, even a small departure from ideal construction of such a resonator may cause substantial loss.

The low-loss regions of Fig. 15 represent three sets of possible geometries:

(1) both mirrors are concave and the centre of curvature of each mirror lies beyond the other mirror;

(2) both mirrors are concave and the centre of curvature of each mirror lies between the other mirror and the centre of curvature of the latter;

(3) one mirror is convex and the other is concave, and the centre of curvature of the concave mirror lies between the convex mirror and the centre of curvature of the latter.

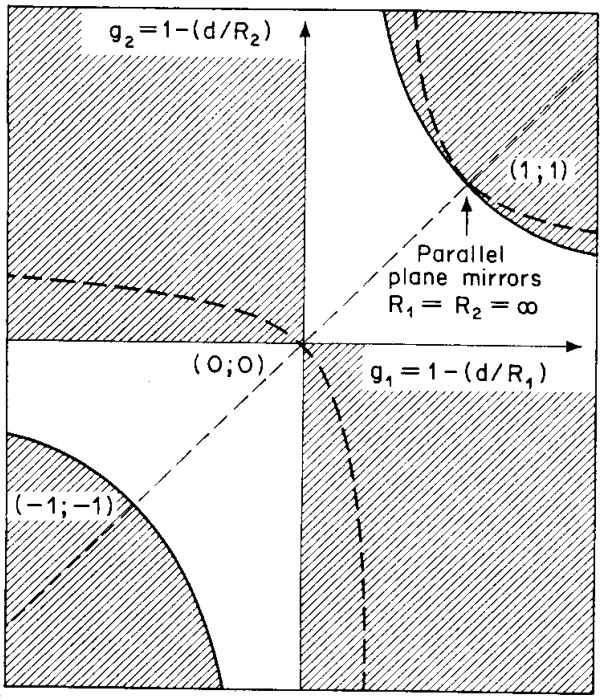


FIG. 15. Stability diagram for various mirror configurations. The shaded regions correspond to high-loss (unstable) systems, and the unshaded regions to stable systems. (After Fox and Li.[24])

Of course, the transition between low-loss and high-loss regions is not as sharp as is suggested by geometrical optics. It is a gradual transition which becomes more and more abrupt as the Fresnel number N increases.[24] In any case, it turns out that mirrors with equal radii confocally spaced give rise to the lowest loss. The loss decreases with increase in the Fresnel number, in the low-loss region, but fluctuates without eventually decreasing, in the high-loss region. The total optical path in an n-fold system is nd, so that the conditions (21), (22) for resonance become

$$nd = q\frac{\lambda}{2} + \varepsilon\frac{\lambda}{4}, \tag{42}$$

where $\varepsilon = 1$ for symmetric modes and $\varepsilon = 0$ for antisymmetric modes.

However, not all the resonances (42) may be present in a real system due to the fact that the system is folded upon itself. Some eigenfunctions may be identically zero, due to destructive interference in successive round trips. For instance, Boyd and Kogelnik[60] give the following formula for the case of rectangular mirrors

$$\frac{2d}{\lambda} = q + \frac{1}{\pi}(2s+\varepsilon)\arccos\sqrt{(g_1 g_2)}, \tag{43}$$

where q and s are positive integers and ε has the same meaning as before. By eliminating $g_1 g_2$ using eqn (41), it is easy to show that all the resonances (43) are included in the general expression (42), whereas the reverse is not true. Of course (43) could be derived from eqn (28).

6. Roof-Mirror Resonators

Stable spherical-mirror resonators, of which the plane parallel Fabry–Pérot and its relatives[61-64] are particular cases, have been most widely investigated. However, there are other stable geometries which may have some special useful property, such as enhanced mode selectivity,[65-66] large mode volume[67] or a highly concentrated field with low loss.[68]

The use of a diffraction grating as a mirror of a resonator has recently found an interesting application in the so called "orotron". This device is very useful for the generation of millimeter waves.[69-71]

Sometimes unstable spherical-mirror resonators can also find useful applications.[72-75] The spill-over radiation can be conveniently used for the extraction of energy (coupling to external space or to other devices). Unstable geometries may also be used to provide large mode volume and good lateral-mode selection.

Another interesting geometry first analysed by the author is that of the flat-roof resonator.[76-77] In this type of resonator (Fig. 16), both terminal mirrors are roofs with extremely small angles (10^{-6} to 10^{-3} radians) which in practice appear to be very nearly flat.

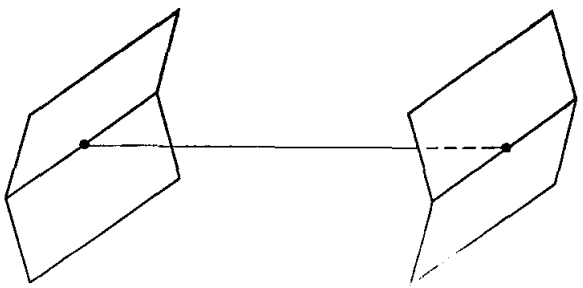


Fig. 16. Roof-mirror resonator. (After Toraldo di Francia.[77])

In order to investigate the modes of the flat-roof resonator, it is expedient to start with a closed diamond cavity GBHE (Fig. 17) of very small angle α. Denoting the distance between the two vertices B and E by d, we will put $d = q_0\lambda/2$, where q_0 represents a very large number, not necessarily an integer.

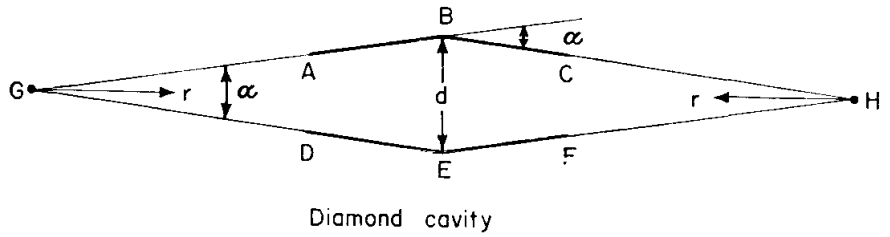


FIG. 17. Showing how a flat roof resonator can be considered as a section of a diamond cavity. The angle α is between 10^{-6} and 10^{-3} radians. (After Toraldo di Francia.[77])

The left hand side of the cavity will be described in terms of cylindrical coordinates r, z, ϕ with the z axis through G, while the right hand side will be described in terms of a symmetrical system of coordinates with the z axis through H. In both halves, solutions of Maxwell's equations with the electric (or magnetic) field parallel to z are given by

$$E = J_\nu(kr)\cos(\nu\phi). \tag{44}$$

with the boundary condition

$$\nu = q\,\frac{\pi}{\alpha}, \tag{45}$$

where q is an integer. We are interested in the case where $q \simeq q_0$ with $q < q_0$. The order of the Bessel function ν then turns out to have a very large value. In the region of interest, i.e. near the centre of the cavity, kr also has a very large value, of the same order of magnitude as ν. We can therefore use the asymptotic expressions for Bessel functions of large order and argument.[78]

It is expedient to consider the "cut-off" plane AD (and correspondingly CF) for which $AD = q\lambda/2$ and to introduce the abscissa x with respect to the cut-off plane, measured positively from the centre of the cavity. We can write

$$J_\nu(kr) \simeq \frac{\cos\left[\dfrac{8\pi}{3}\left(\dfrac{\alpha|x|^3}{q\lambda^3}\right)^{\frac{1}{2}} - \dfrac{\pi}{4}\right]}{\pi\left(\dfrac{q|x|}{\alpha\lambda}\right)^{\frac{1}{4}}} \qquad \text{for } x < 0 \tag{46}$$

and

$$J_\nu(kr) \simeq \frac{\exp\left[-\frac{8\pi}{3}\left(\frac{\alpha x^3}{q\lambda^3}\right)^{\frac{1}{2}}\right]}{2\pi\left(\frac{qx}{\alpha\lambda}\right)^{\frac{1}{4}}} \qquad \text{for } x > 0. \tag{47}$$

Equation (46) represents a standing wave. We must use the fact that α is very small and ignore the curvature of the nodal and antinodal surfaces. It is then an easy matter to match the fields in the left and right halves of the cavity. The resonance condition for the mth mode then becomes

$$\lambda = \frac{2d}{q}\left\{1 - \frac{1}{2}\left[\frac{3\alpha(2m+1)}{4q}\right]^{2/3}\right\}. \tag{48}$$

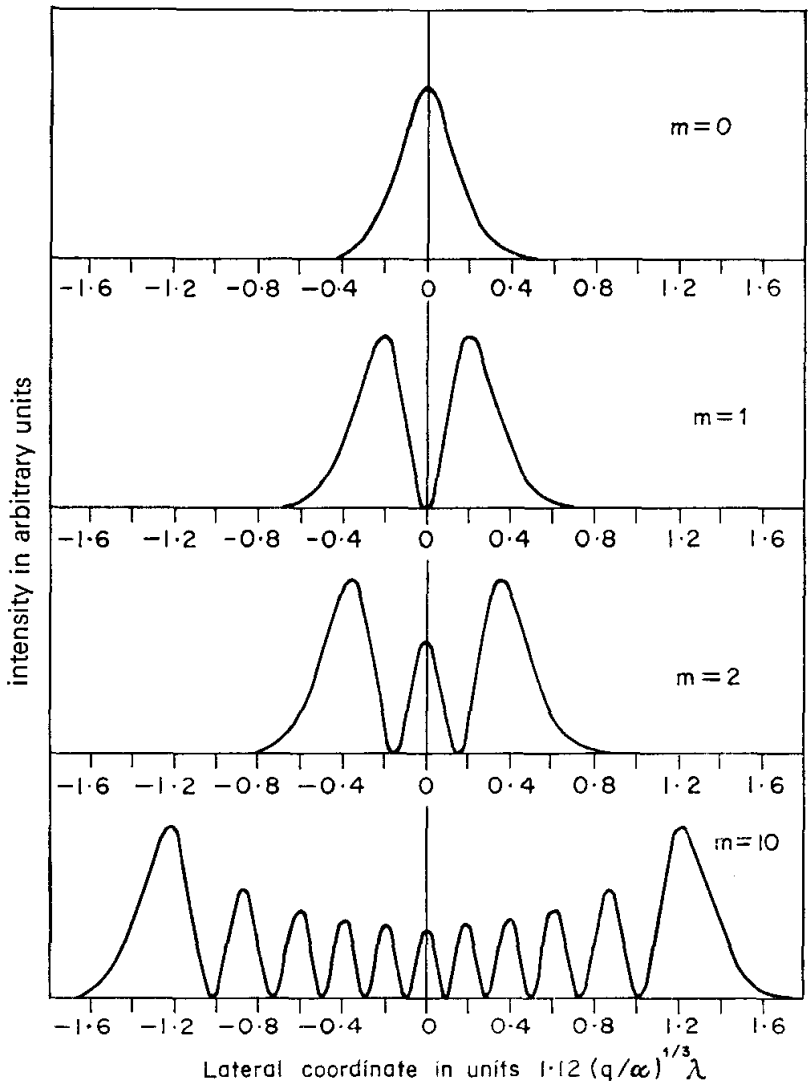


FIG. 18. Intensity distributions for several modes in a flat roof resonator. (After Toraldo di Francia.[77])

Equation (47) shows that outside the cut-off planes the field falls off exponentially. It is therefore permissible to limit the walls of the cavity to the portions *ABC*, *DEF*, thus obtaining a flat-roof resonator.

Figure 18 shows the intensity distribution of a few modes, calculated using the above formulae.

This type of resonator and its optically equivalent beam waveguide, consisting of a sequence of special narrow-angle prisms have been investigated by Kahn[79] in terms of geometrical optics. Kahn has calculated the transverse field distribution using simple electromagnetic theory. An appropriate definition of the spot size is found to be in agreement with that of Toraldo di Francia.[77]

The results of the approximate theoretical approach (valid for very small values of α), have been verified by Checcacci, Consortini and Scheggi[80-82] by numerical computation and by experiment in the X band.

The numerical solution of the integral equation for the flat-roof was found by the Fox and Li method of iteration. The losses were evaluated as functions of the roof angle α. The investigation was not limited to small values of α. Negative values corresponding to convex flat-roof resonators were also considered. Figure 19 shows the power losses per transit plotted versus α for $N = 2{\cdot}22$. In the positive region of α the losses for the zeroth order mode have minima and maxima. In the region of the first maximum, the second order

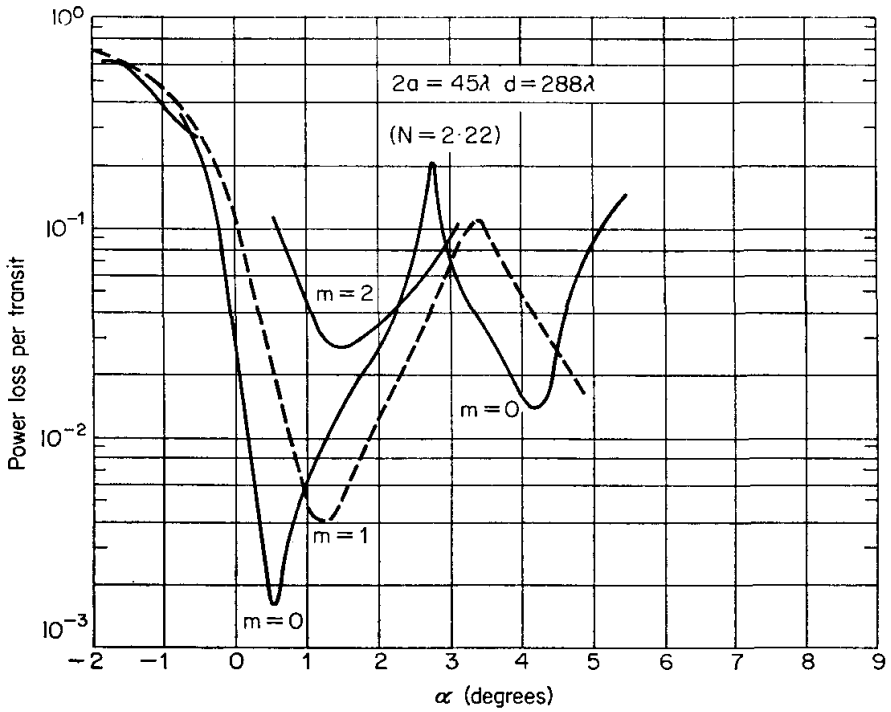


FIG. 19. Plot of the power loss per transit versus the angle α in flat roof resonators. (After Checcacci *et al.*[86])

mode losses have a broad minimum. Thus there is a region which is unstable for the fundamental mode, but in which a higher order mode has lower losses.

The existence of minimum losses corresponding to particular values of α had been found by Voitovich[83] for a resonator terminated by conical reflectors.

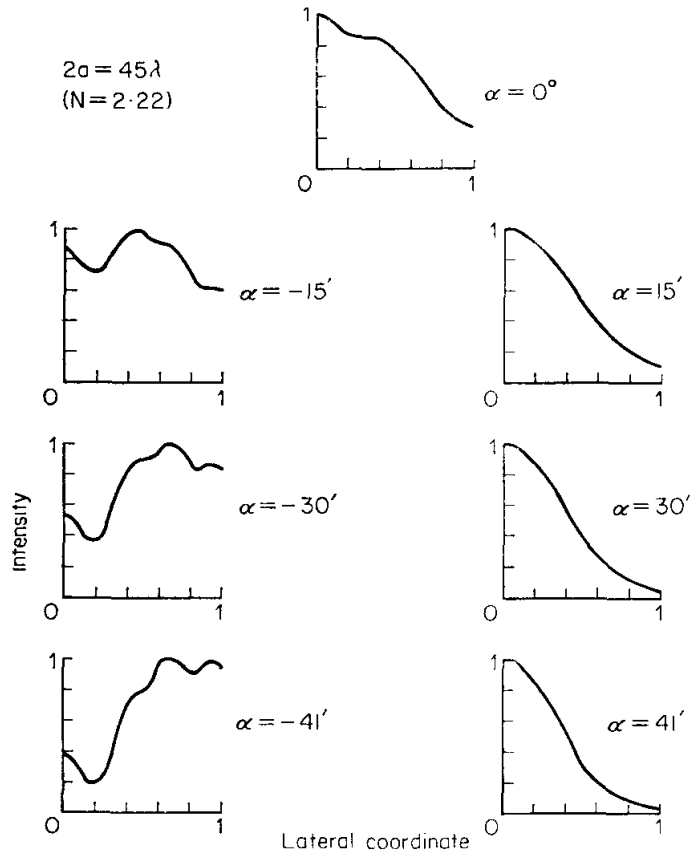


FIG. 20. Zeroth order mode patterns for positive and negative values of α. (After Checcacci *et al.*[86])

For negative values of α, the losses increase with $|\alpha|$ as one would expect due to the geometrical instability .There is also a region here where an inversion occurs between the fundamental and higher order modes. In passing from a convergent to a divergent resonator the mode shapes change noticeably as can be seen in Fig. 20 where zeroth order mode patterns are shown for some positive and negative values of α. It can be observed that the convex flat-roof resonator has a larger mode volume than the geometrically stable concave resonator of the same type. The fact that the field intensity tends to shift its maxima towards the edges of the mirrors by increasing $|\alpha|$ can be very useful in resonator coupling.

Experimental tests were carried out in the X band using a roof mirror of variable angle. Figure 21 shows a comparison between the approximate

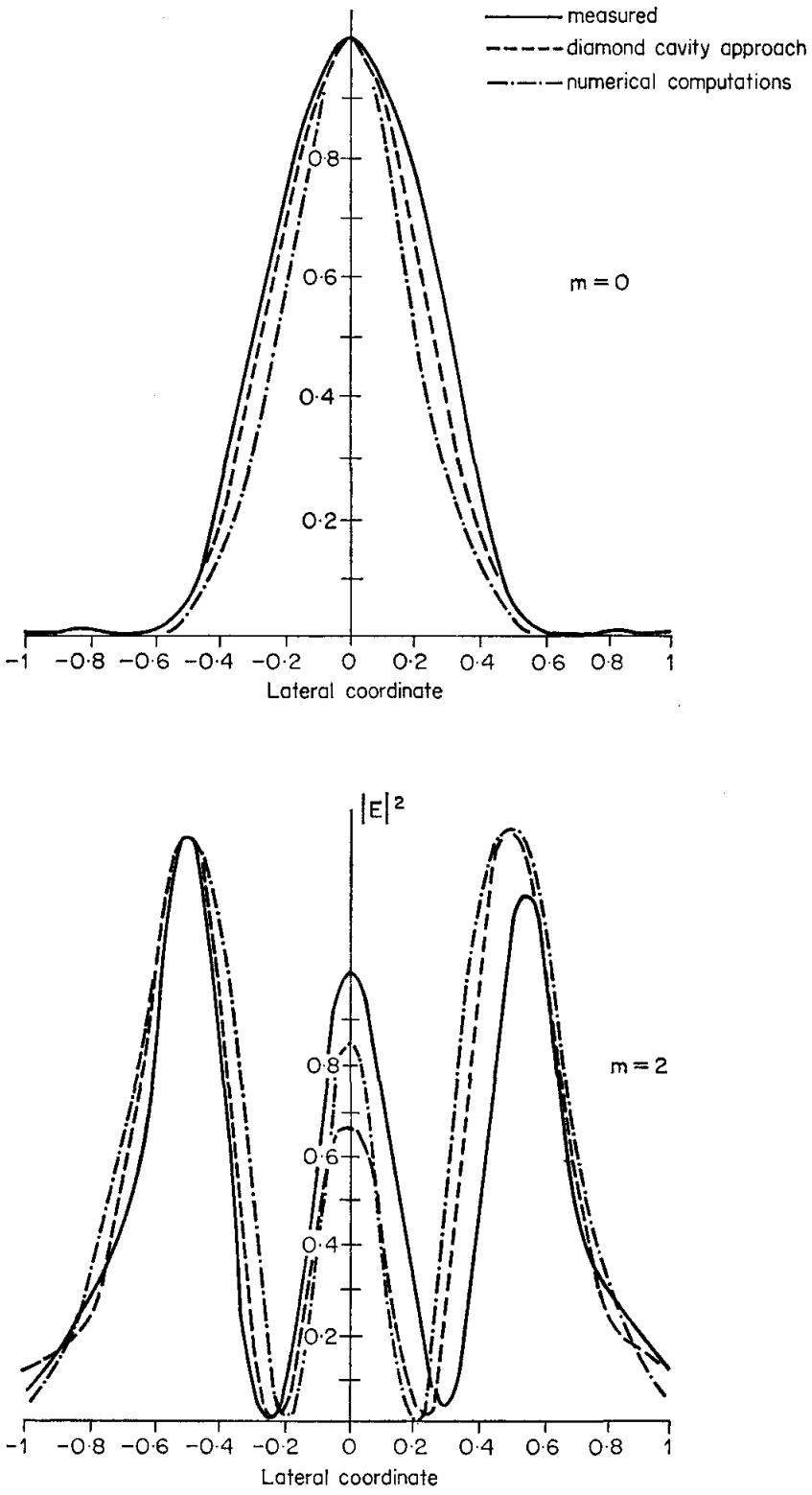


FIG. 21. Comparing the calculated field distributions, in the zeroth and second order modes, with experimental values. Mirror aperture $2a = 45\lambda$ and $\alpha = 1° 16'$.

theory, the iterative computation and the experimental measurements for the field distribution (intensity) of the first two even (symmetric) modes.

The problem of modes and losses in a 90°-roof resonator has been tackled by Checcacci and Scheggi.[84, 85] They carried out some experimental tests on an X-band model consisting of a plane mirror and a right angle roof mirror. In order to investigate the effect of the departure of the roof angle from 90°, this was varied over a small range around 90°.

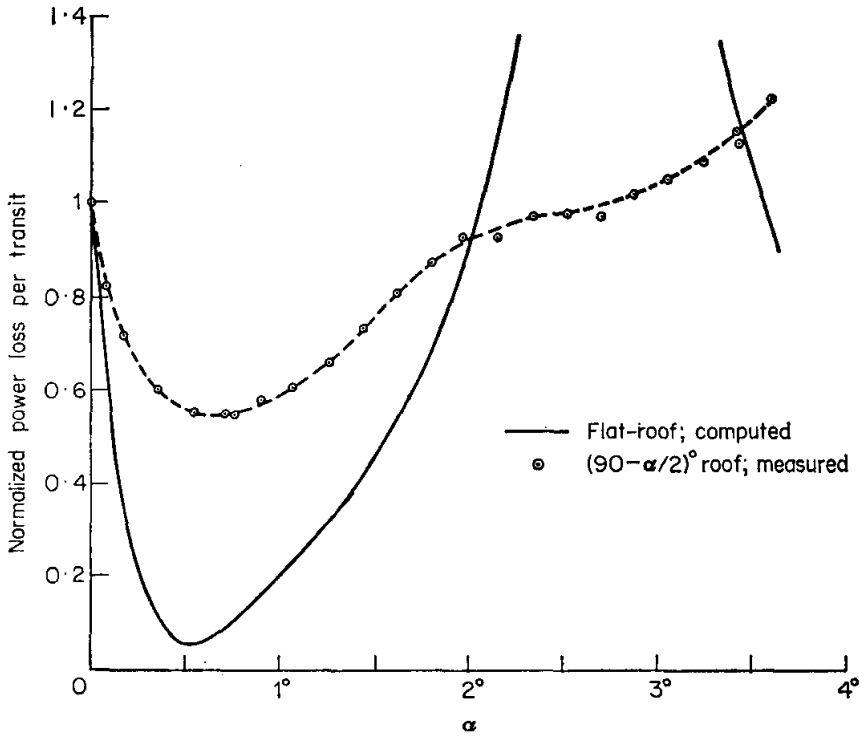


FIG. 22. Comparing the power loss of the $(90-\alpha/2)°$ roof resonator with that of the flat roof resonator. (After Checcacci and Scheggi.[84])

In the geometrical optics approximation, the behaviour of a $(90-\alpha/2)°$ roof resonator may be predicted from that of an equivalent flat-roof, at least for small values of α. For $\alpha = 0$ the flat-roof becomes a plane mirror, hence one can expect that the 90°-roof mirror will also be equivalent to a plane mirror. The behaviour of the energy loss per transit as derived from the measured Q's is plotted versus α in Fig. 22 along with the analogous curve computed for the equivalent flat-roof mirror. For small values of α ($<2°$) the two curves show vaguely similar behaviour, but the similarity disappears for larger values of α and in particular the experimental curve does not follow the high loss region of the flat-roof resonator.

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Review of Nonlinear Optical Phenomena in Condensed Matter

N. BLOEMBERGEN

1. INTRODUCTION

This chapter will review a variety of nonlinear optical phenomena which have become accessible to experimental investigation in the past decade due to the extremely high light intensities obtainable in pulsed laser beams. Since condensed systems with a large number of degrees of freedom can exhibit a complex array of different phenomena, many simplifying assumptions will be made in the theory. From the outset the electromagnetic fields will be treated classically. The chapters by Glauber, Haken and Louisell in this volume show that a complete quantum mechanical treatment of some of the simpler nonlinear phenomena, such as parametric generation, already exists.[1] The semiclassical approach permits a quicker, unified survey and leads to good agreement with practically all experimental situations, provided spontaneous emission noise fields are added in an *ad hoc* manner.

The general Hamiltonian may be divided into the vacuum field, the material Hamiltonian and a term representing the interaction between electromagnetic radiation and matter. In addition, the interactions with a thermal reservoir with a large number of degrees of freedom will lead to a phenomenological damping term,

$$\mathscr{H} = \mathscr{H}_{\mathrm{EM}} + \mathscr{H}_{\mathrm{mat}} + \mathscr{H}_{\mathrm{int}} + \mathscr{H}_{\mathrm{damping}} \tag{1}$$

In general $\mathscr{H}_{\mathrm{EM}}$ contains all the vacuum oscillators, and $\mathscr{H}_{\mathrm{mat}}$ in diagonalized form contains all the energy levels of the condensed matter under consideration. In many situations it is possible to restrict the attention to a limited number of electromagnetic oscillators and/or a limited number of material energy levels. The interaction Hamiltonian may often by written, e.g. for bound electrons interacting with light waves or electric fields of lower frequency, in the electric dipole approximation,

$$\mathscr{H}_{\mathrm{int}} = -\sum_i e_i \mathbf{r}_i \cdot \mathbf{E} = -\sum_i \boldsymbol{\mu}_i \cdot \mathbf{E}, \tag{2}$$

where μ_i is the electric dipole moment operator of the ith particle.

For a statistical ensemble of particles, the density matrix ρ describes the

quantum mechanical behaviour of the material system. In the absence of damping it obeys the operator equation

$$i\hbar\dot{\rho} = [\mathscr{H}_0, \rho] - \mathbf{E} \cdot [\boldsymbol{\mu}, \rho]. \tag{3}$$

The macroscopic electric polarization of the medium is given by

$$\mathbf{P} = N \operatorname{Tr} (\boldsymbol{\mu}\rho), \tag{4}$$

where N is the number of identical particles per unit volume. The electromagnetic field obeys the classical Maxwell equations. The wave equation for the electric field then takes the form

$$\nabla \wedge \nabla \wedge \mathbf{E} + \frac{1}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} = -\frac{4\pi}{c^2} \frac{\partial^2 \mathbf{P}}{\partial t^2}. \tag{5}$$

The set of equations (3–5) constitute the starting point of the semiclassical treatment of nonlinear optics. Only the simplest case allows solutions in a closed form. As an example, the propagation of a single quasi-monochromatic pulsed wave travelling through a medium containing atomic systems with only two energy levels will be discussed in the next paragraph. For more complicated situations one must resort to perturbation procedures.

2. Light Pulse Propagation Through a Two Level System

When a laser pulse, e.g. from a ruby laser, is passed through a second ruby crystal, it will normally be absorbed, if the initial population of the two levels of the Cr^{3+} ion involved in the laser transition is normal. If the second ruby laser crystal is also optically pumped to yield an inverted population, the laser pulse will be amplified. For very intense light pulses there may, however, be no net gain or loss of light energy. This effect is called self-induced transparency, and was first demonstrated and discussed in detail by McCall and Hahn.[2] An excellent detailed account, consolidating the efforts of many other authors, has recently been given by Icsevgi and Lamb.[3] The case of one electromagnetic mode interacting with a two level system provides the simplest physically meaningful situation of nonlinear response.

The electric field is assumed to be a quasi-monochromatic travelling wave, linearly polarized in the x-direction, with a slowly varying amplitude and phase

$$E_x = A(z,t) \exp (ikz - i\omega t) = \mathscr{E}(z,t) \exp [i\phi(z,t)] \exp (ikz - i\omega t). \tag{6}$$

The two energy levels are denoted by $|a\rangle$ and $|b\rangle$, with energies $-\frac{1}{2}\hbar\omega_0$ and $+\frac{1}{2}\hbar\omega_0$ respectively. The only nonvanishing matrix elements of the electric dipole moment operator of interest are

$$(\mu_x)_{ab} = (\mu_x)_{ba} = \mu.$$

The frequency ω is chosen close to the resonant frequency ω_0 with $\Delta = \omega_0 - \omega$. The wave number $k = \omega c^{-1}\varepsilon^{1/2}$ includes the effect of the linear dielectric constant of the bulk crystal, exclusive of the two Cr^{3+} levels. It may also include the effect of other light losses by giving the linear dielectric constant an imaginary part.

It is well known that the equation of motion of a two by two density matrix has a one to one correspondence with the motion of a real classical magnetization vector. Introduce three real components, (X, Y, Z) which are slowly varying quantities in the interaction representation, such that;

$$X - iY = 2\rho_{ab} \exp(-ikz + i\omega t)$$
$$Z = \rho_{aa} - \rho_{bb}$$
$$1 = \rho_{aa} + \rho_{bb}, \quad \rho_{ab} = \rho_{ba}^{*}.$$

In terms of these quantities the operator equation of motion (3) may be written as

$$\dot{X}(z, t, \Delta) = -\Delta Y(z, t, \Delta) - \frac{1}{T_2} X, \tag{7}$$

$$\dot{Y}(z, t, \Delta) = \Delta X(z, t, \Delta) + \hbar^{-1} \mu \mathscr{E}(z, t)\, Z(z, t, \Delta) - \frac{1}{T_2} Y, \tag{8}$$

$$\dot{Z}(z, t, \Delta) = -\hbar^{-1} \mu \mathscr{E}(z, t)\, Y(z, t, \Delta) - \frac{1}{T_1}(Z - Z_0). \tag{9}$$

The last terms on the right-hand side are phenomenological damping terms. In writing these equations, rapidly varying components in X, Y and Z have been ignored. These terms would give rise to harmonics of small amplitude, as discussed in the next section. The truncation of these counter-rotating terms permits us to restrict ourselves to one mode of the electromagnetic field. The equations of motion would be strictly correct for a circularly polarized wave and magnetic dipole matrix elements of the form $(\mu_x)_{ab} = -i(\mu_y)_{ab}$. The electric polarization is given by

$$P_x(z, t, \Delta) = N\mu[X(z, t, \Delta) - iY(z, t, \Delta)] \exp(ikz - i\omega t).$$

It is seen that X corresponds to the in-phase component and Y to the component of polarization which is 90° out of phase with the electric field. Substitution of this polarization into the wave equation (5) leads to the complex amplitude equation,

$$-ik \frac{c^2}{n^2} \frac{\partial A}{\partial z} - i\omega \frac{\partial A}{\partial t} = \frac{2\pi N \mu \omega^2}{n^2} (X - iY).$$

Here n is the linear index of refraction of the crystal, exclusive of the two level system. Second order derivatives in A have been ignored (slowly varying

envelope approximation). Equating the real and imaginary parts on both sides and allowing for a distribution of resonant frequencies $g(\Delta)$ (inhomogeneous broadening), one finds

$$\frac{c}{n}\frac{\partial \mathscr{E}(z,t)}{\partial z} + \frac{\partial \mathscr{E}(z,t)}{\partial t} = \frac{2\pi N \mu \omega}{n^2}\int_{-\infty}^{+\infty} Y(z,t,\Delta)\, g(\Delta)\, \mathrm{d}\Delta \tag{10}$$

and

$$\mathscr{E}\left(\frac{c}{n}\frac{\partial \phi}{\partial z} + \frac{\partial \phi}{\partial t}\right) = \frac{2\pi N \mu \omega}{n^2}\int_{-\infty}^{+\infty} X(z,t,\Delta)\, g(\Delta)\, \mathrm{d}\Delta. \tag{11}$$

The solutions of the set of five coupled partial differential equations with five real variables have been discussed by a large number of authors.[2,3] Icsevgi and Lamb[3] add a sixth equation, because $\rho_{aa}+\rho_{bb}$ is not assumed to be equal to unity. If the laser transition is between two excited levels, each of which may be populated independently by a pump mechanism and may decay (relax) independently to lower states, this is a useful extension. So is the generalization to moving particles, where the density matrix ρ also depends on the velocity. For the propagation characteristics of a travelling wave the resultant Doppler effect may be incorporated in the inhomogeneous broadening $g(\Delta)$.

Here we shall restrict ourselves to a solution, in which the damping terms in eqns (7–9) may be ignored. This solution applies in the limit $\hbar^{-1}\mu\mathscr{E}T_2 \gg 1$. For the case of exact resonance and no inhomogeneous broadening, $g(\Delta) = \delta(\Delta)$ and $\Delta = 0$, the equations have a solution with $X = 0$ and $\phi = 0$. The remaining set of three coupled equations for $\mathscr{E}, Y$ and Z can be solved exactly, with these quantities being functions of the argument $t-z/v$. They correspond to the solution in which the envelope propagates without change of shape with a velocity v. If $g(\Delta)$ is an even function of Δ, one can still find solutions with $\phi = 0$, while $Y(\Delta)$ is an even function of Δ and $X(\Delta)$ an odd function. Several workers[4] have found periodic solutions in terms of Jacobian elliptic functions, whereas McCall and Hahn had shown earlier that an aperiodic pulse solution exists in the form

$$\mathscr{E}(z,t) = \mathscr{E}_0 \operatorname{sech}\left[\hbar^{-1}\mu\mathscr{E}_0\,(t-z/v)\right].$$

At resonance, $\Delta = 0$, the vector $(X = 0)$, Y, Z is rotated through an angle

$$\Phi(z,t) = \int_{-\infty}^{t} \hbar^{-1}\mu\mathscr{E}(z,t')\,\mathrm{d}t'.$$

At the end of the pulse this angle is 2π and the material has returned to its initial state. At high power levels the pulse thus passes without attenuation or gain. The feature which distinguishes it experimentally from pulse propagation

through a saturated medium is that the velocity of propagation is changed,

$$\frac{1}{v} = \frac{n}{c} + \frac{2\pi N\omega\mu^2\tau^2}{\hbar cn}\int_{-\infty}^{+\infty}\frac{g(\Delta)\,d\Delta}{1+(\Delta\omega\tau)^2},$$

where $\tau^{-1} = \hbar^{-1}\mu\mathscr{E}$

If the medium is initially absorbing, energy is taken up by the medium at the beginning of the pulse, and is returned to it at the end of the pulse. This has the effect of a retardation on the propagation of the envelope of constant shape. Another way of expressing this is to say that a fraction of the energy is stored in the medium and this part does not contribute to the energy flux, so that $v \times$ (EM + material energy density) $= (c/n) \times$ EM energy density. This pulse delay has been observed experimentally by McCall and Hahn. The pulse from a ruby laser was passed through a second passive ruby crystal kept at liquid helium temperature to keep T_2 long. For $\hbar^{-1}\mathscr{E}\mu T_2 \gg 1$ transparency occurred.

When the material is initially in an excited state, i.e. behaves as an active laser medium, the pulse shape would tend to propagate with a velocity larger than the velocity of light. This kind of effect has been observed in a laser pulse amplifier. There is no conflict with special relativity theory. There will never be a signal before a time $t - zn/c$, if there were no input before t at $z = 0$. If there is an input, the leading edge is amplified and the shape of the pulse appears to move forward, although it is *not* a stationary or distortionless solution in the strict sense. The hyperbolic secant is of course not a physically realistic input, since it starts at $-\infty$.

Detailed analysis with the aid of computers shows how, in the presence of damping, a large input pulse of arbitrary shape, may break up into a number of 2π pulses. The only stable solutions are of course those in which the material eventually returns to a passive state. For an initially inverted population one would have the emission of π, 3π etc. pulses. For further details references 2–4 should be consulted.

We next turn to the numerous situations, in which the damping of the matrix elements is sufficiently large, or where the frequencies of the applied fields are sufficiently far removed from atomic resonant frequencies, so that the equation of motion of the density matrix may be solved by a perturbation procedure. In this case one need not restrict oneself to a two level system. The response of the material can be described by a series of nonlinear susceptibilities of increasing order.

3. Lowest Order Nonlinear Susceptibility

When $|\hbar^{-1}\mu E| \ll |\Delta + iT_2^{-1}|$, i.e. when the matrix elements of the interaction Hamiltonian measured in frequency units are small compared to the inverse

damping time or well removed from resonance, an iterative perturbation procedure may be used to describe the response of the material to periodic perturbations.[5]

The response of the material to first or linear approximation is given by

$$-i\omega\,\rho_{kl}^{(1)} = -i\omega_{kl}\,\rho_{kl}^{(1)} - \Gamma_{kl}\,\rho_{kl}^{(1)} - i\hbar^{-1}\,[-\boldsymbol{\mu}\,.\,\mathbf{E}(\omega),\,\rho^{0}]_{kl},$$

where $E(\omega)$ is the electric field Fourier component at frequency ω; ρ^0 is the steady state or zero order density matrix, and usually contains only diagonal elements $\rho_{kk}{}^0$ giving the fractional occupation of each state. To second approximation one finds, for example, elements varying at the harmonic frequency 2ω,

$$-2i\omega\,\rho_{kl}^{(2)}(2\omega) = -i\omega_{kl}\,\rho_{kl}^{(2)}(2\omega) - \Gamma_{kl}\,\rho_{kl}^{(2)} - i\hbar^{-1}\,[-\boldsymbol{\mu}\,.\,\mathbf{E}(\omega),\,\rho^{(1)}(\omega)]_{kl}. \quad (12)$$

With the trace relation equation (4), one finds a harmonic polarization

$$P_i(2\omega) = N\,\mathrm{Tr}\,\mu_i\rho(2\omega) = \chi_{ijk}\,(-2\omega,\,\omega,\,\omega)\,E_j(\omega)\,E_k(\omega)\exp\,(-2i\omega t). \quad (13)$$

The nonlinear susceptibility tensor describing a polarization quadratic in the electric field amplitudes is thus determined by this procedure. In the case of negligible absorption at both the fundamental and the second harmonic frequency and in the case where μ_x has no diagonal elements (no permanent dipoles), one finds, for example.

$$\begin{aligned}\chi_{xxx}\,(-2\omega,\,\omega,\,\omega) = \sum_{g\neq}\sum_{n}\sum_{\neq n'} Ne^3\,\mu_{x,gn}\,\mu_{x,nn'}\,\mu_{x,n'g}\,\rho_{gg}(0)\,\hbar^{-2}\\ \times[(2\omega-\omega_{ng})^{-1}\,(\omega-\omega_{n'g})^{-1}+(2\omega+\omega_{ng})^{-1}\,(\omega+\omega_{n'g})^{-1}\\ -(\omega+\omega_{ng})^{-1}\,(\omega-\omega_{n'g})^{-1}] \quad (14)\end{aligned}$$

More generally the incident electromagnetic field may consist of a number of running waves

$$\mathbf{E}(\mathbf{r},t) = \sum_n [A_n(\omega_n)\exp(i\mathbf{k}_n\,.\,\mathbf{r}-i\omega_n t) + A_n{}^*(\omega_n)\exp\,(-i\mathbf{k}_n\,.\,\mathbf{r}+i\omega_n t)].$$

The Cartesian component in the i-direction of a polarization at the sum frequency, proportional to an electric field at ω_1 and an electric field at ω_2 may then be written as,

$$\begin{aligned}P_i^{NL}(\mathbf{r},\omega_1+\omega_2) = D\,\chi_{ijk}(-\omega_1-\omega_2,\,\omega_1,\,\omega_2)\\ \times E_j(\omega_1)\,E_k(\omega_2)\exp\,[i(\mathbf{k}_1+\mathbf{k}_2)\,.\,\mathbf{r}] \quad (15)\end{aligned}$$

The real polarization is obtained by multiplying by the time factor $\exp\{-i(\omega_1+\omega_2)t\}$ and adding the complex conjugate.

The reality of the physical polarization requires

$$\chi_{ijk}(-\omega_1-\omega_2,\,\omega_1,\,\omega_2) = \chi^*{}_{ijk}\,(\omega_1+\omega_2,\,-\omega_1,\,-\omega_2).$$

If the material is not dissipative, the nonlinear susceptibility should be invariant for time reversal, which is equivalent to replacing each frequency by its negative value. The result is that χ_{ijk} must be real. This follows of course also from the microscopic calculation in the absence of damping. The summation convention over the repeated indices j and k is understood in eqn (15). Each index runs separately over all three coordinates, x, y, and z. Because the order of the field components on the right hand side is of no importance, one has obviously

$$\chi_{ijk}(-\omega_1-\omega_2, \omega_1, \omega_2) = \chi_{ikj}(-\omega_1-\omega_2, \omega_2, \omega_1).$$

D is the degeneracy factor, describing the number of distinguishable permutations of the frequencies; $D = 2$, if $\omega_1 \neq \omega_2$. Eqn (15) may also be used to describe the second harmonic polarization with $D = 1$ and $\omega_1 = \omega_2$. The same expression may also be used to describe the polarization at the difference frequency $\omega_1-\omega_2$, by simply taking one positive and one negative frequency Fourier component.

Explicitly, we have

$$P_i^{NL}(\omega_1-\omega_2) = (D = 2)\,\chi_{ijk}(-\omega_1+\omega_2, \omega_1, -\omega_2)\,E_j(\omega_1)\,E_k^*(\omega_2). \quad (16)$$

The rectification of light is also described by taking the two frequencies equal and opposite. A dc polarization is induced proportional to the intensity of the light,

$$P_i^{NL}(0) = (D = 2)\chi_{ijk}(0, \omega_1, -\omega_1)\,E_j(\omega_1)\,E_k(-\omega_1) + (D = 2)\chi_{ijk}(0, \omega_2, -\omega_2)\,E_j(\omega_2)\,E_k(-\omega_2). \quad (17)$$

Note that the degeneracy factor remains two in this case, because positive and negative frequencies are distinguishable.

The nonlinear susceptibility tensor is obviously a third rank tensor, connecting three polar vectors. The explicit forms for the 32 crystal classes have been given by Butcher and Giordmaine[6]. All elements must of course vanish, if there is a centre of inversion. In the general case of the triclinic class 1 there are 27 independent elements, because the tensor need not be symmetric in any pair of indices, if all frequencies are different. For second harmonic generation the elements are obviously symmetric in the indices j and k. Therefore χ_{ijk} $(-2\omega, \omega, \omega,)$ has the same symmetry properties as the piezoelectric tensor, which connects the polar vector $\mathbf{P}$ with the symmetric stress tensor.

The tensor χ_{ijk} $(-\omega_1-\omega_2, \omega_1, \omega_2)$ has a part which is antisymmetric in the last pair of indices for $\omega_1 \neq \omega_2$ and this part is explicitly displayed in the tables of Giordmaine which also include textures, such as liquid crystals and isotropic liquids. Because of this antisymmetric part sum frequency generation is possible in the cubic class 432, which is not piezoelectric, and does not allow second harmonic generation.

Sum frequency generation is also possible in an isotropic fluid which does not possess a centre of inversion. This fluid must show natural optical activity. It must contain a non-racemic mixture of optically active molecules. Sum frequency generation has been demonstrated in a solution of *l*- or *d*- arabinose.[7]

In an isotropic fluid the third rank tensor must of course reduce to a scalar in a simple vector relation. The sum frequency generation can be described in this case by

$$\mathbf{P}(\omega_1+\omega_2) = 2\chi_{xyz}\{\mathbf{E}(\omega_1)\wedge\mathbf{E}(\omega_2)\}.$$

Here χ_{xyz} may be considered as a pseudo-scalar property of the fluid. For the coherent radiation from this polarization to be observable, it is necessary that the light beams at ω_1 and ω_2 propagate at an angle to each other.

In a non-dissipative medium a thermodynamic time averaged potential may be defined, from which the nonlinear polarization is derived by differentiation. This leads to the permutation symmetry relation,

$$D'\chi_{ijk}(-\omega_1-\omega_2, \omega_1, \omega_2) = \chi_{jik}(\omega_1, -\omega_1-\omega_2, \omega_2),$$

which states that any pair of indices in the susceptibility tensor may be interchanged, provided the corresponding frequencies are also permuted. $D' = 1+\delta_{jk}\delta_{12}$ is another degeneracy factor. It is usually unity, but it equals two, if the frequencies ω_1 and ω_2 are equal and $j = k$. A nontrivial application of this relation is the prediction that the rectification of light according to eqn (17) is described by the same constant that describes the linear Kerr or Pockels effect. Both are derived from

$$U^{NL} = \chi_{ijk}(0, \omega_1, -\omega_1)\, E_{idc}(0)\, E_j(\omega_1)\, E_k^*(\omega_1).$$

Note that $4\pi\chi_{ijk}E_{idc}$ gives the part of the dielectric constant ε_{jk} which is linear in the applied dc field. This prediction has been confirmed experimentally.[8]

In the literature much attention has been paid to the Kleinman symmetry relation or Kleinman conjecture. Kleinman[9] notes that in many experiments the frequencies all lie in a range of optical transparency, where dispersive effects are small. If dispersive effects are ignored, because the absorption bands in the ultraviolet and infrared are sufficiently far removed from the optical frequencies involved, the susceptibility tensor becomes, of course, independent of the frequencies and should become symmetric for the interchange of any two indices. This leads to the vanishing of additional elements of the tensor in certain cases.

In the point group 32, for example, general symmetry requires that

$$\chi_{xyz}(-2\omega, \omega, \omega) = \chi_{yxz}(-2\omega, \omega, \omega), \quad \text{and} \quad \chi_{zxy}(-2\omega, \omega, \omega) = 0,$$

if z denotes the trigonal axis. If the Kleinman symmetry is invoked the first two elements should also vanish. Experimentally[10] an upper limit for these elements of second harmonic generation has been set in quartz, which is less than 2% of the element χ_{xxx}. Other examples [10] of the Kleinman symmetry relation are the equality, within the experimental error, of the elements $\chi_{xyz}(-2\omega, \omega, \omega)$ and $\chi_{zxy}(-2\omega, \omega, \omega)$ in KDP, which belongs to the tetragonal class $\bar{4}2m$, and of $\chi_{zxx}(-2\omega, \omega, \omega)$ and $\chi_{xzx}(-2\omega, \omega, \omega)$ in $BaTiO_3$ which belongs to $4mm$. These are illustrations of the fact that the dispersion of the optical nonlinearity is indeed small for these crystals in the visible and near ultra-violet.

For the same reason the antisymmetric part describing sum frequency generation, which vanishes for second harmonic generation, may be expected to be small, because it exists only to the extent that dispersion is not negligible. In general, such elements may be expected to be only a few percent of the other elements, except in regions of strong dispersion. In these regions dissipative processes are usually also important and the nonlinear susceptibilities become complex.

An interesting application of symmetry considerations results if circularly polarized light is propagating along an axis of symmetry of the crystal.[11] Since the combined system of crystal and electromagnetic field has this symmetry axis, the resultant harmonic polarization must also be circularly polarized or vanish. It has been shown that the second harmonic wave generated by a circularly polarized fundamental wave propagating along a threefold axis is circularly polarized in the opposite sense. If this fundamental beam propagates along a fourfold axis, the harmonic intensity vanishes. The interesting question of conservation of angular momentum in these situations has been discussed in terms of the effect of the trigonal crystalline field potential on the electronic wave functions.[11]

4. Nonlinear Coupling between Electromagnetic Waves

The nonlinear polarization must now be substituted back into Maxwell's equations. For a wave at the sum frequency $\omega_3 = \omega_1 + \omega_2$ one obtains for example,

$$\nabla \wedge \nabla \wedge \mathbf{E}_3 - (\omega_3^2/c^2)\,\varepsilon(\omega_3)\,.\,\mathbf{E} = 4\pi(\omega_3^2/c^2)\,\chi^{NL}(-\omega_1-\omega_2, \omega_1, \omega_2) : \mathbf{E}_1\mathbf{E}_2. \tag{18}$$

This equation describes the generation of the sum frequency, if the fields $\mathbf{E}_1$ and $\mathbf{E}_2$ can be considered as constant, given paramenters. The equation has been solved for a variety of boundary conditions and geometrical configurations.

If the amplitude of the field $\mathbf{E}_3$ becomes appreciable, the nonlinearities produced by this field must in turn be taken into account. This leads to a coupling between electromagnetic waves, which would have propagated independently a in linear medium. The response of the medium is hidden in a coupling coefficient, which is proportional to an element of the nonlinear susceptibility. For many experimental situations this is precisely what is desired, because only the electromagnetic fields are detected by the observer.

The lowest order nonlinear susceptibility,

$$\chi^{NL}_{321} = \hat{a}_3^* \cdot \chi(-\omega_3, \omega_2, \omega_1) : \hat{a}_2\, \hat{a}_1,$$

leads to a coupling between three electromagnetic waves, with polarization directions $\hat{a}_3$, $\hat{a}_2$, and $\hat{a}_1$ and frequencies $\omega_3 = \omega_1 + \omega_2$, ω_2 and ω_1 respectively. The momentum vector mismatch is $\Delta k\hat{z} = \mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}_3$ occurring in the $\hat{z}$-direction, normal to the boundary of the nonlinear medium. The three coupled wave equations reduce to three coupled equations for the slowly varying complex amplitudes,

$$\frac{\partial A_1}{\partial z} + \frac{1}{v_{g_1}} \frac{\partial A_1}{\partial t} = 2\pi i\, \chi^{NL}_{321}\, (\omega_1/n_1 c) A_3 A_2^* \exp(-i\Delta k z) \tag{19a}$$

$$\frac{\partial A_2}{\partial z} + \frac{1}{v_{g_2}} \frac{\partial A_2}{\partial t} = 2\pi i\, \chi^{NL}_{321}\, (\omega_2/n_2 c) A_3 A_1^* \exp(-i\Delta k z) \tag{19b}$$

$$\frac{\partial A_3}{\partial z} + \frac{1}{v_{g_3}} \frac{\partial A_3}{\partial t} = 2\pi i\, \chi^{NL}_{321}\, (\omega_3/n_3 c)\, A_1 A_2 \exp(+i\Delta k z) \tag{19c}$$

where v_{g_1}, v_{g_2}, and v_{g_3} are the group velocities of the three envelopes. Use has been made of the permutation symmetry relation.

These equations have the following integrals

$$\frac{n_1 |A_1|^2}{\omega_1} + \frac{n_3 |A_3|^2}{\omega_3} = \text{constant}$$

$$\frac{n_2 |A_2|^2}{\omega_2} + \frac{n_3 |A_3|^2}{\omega_3} = \text{constant}'.$$

They express the fact that if the photon flux density in the wave at the sum frequency is increased by a certain amount, the photon flux densities in the waves at ω_1 and ω_2 are decreased by the same amount. These equations are known as the Manley–Rowe relations in parametric amplifier theory. The remaining relations of the three coupled complex amplitude equations can be reduced to two real equations for the amplitude $|A_3|$ and the phase difference

$\phi_3-\phi_1-\phi_2$. Exact solutions in terms of elliptic functions have been given. These solutions are very similar in form to the solutions more recently obtained for the pulse propagation, discussed in Section 2. The set of equations (19a–c) displays of course striking similarity with the set of equations (7–11). We refer to appendix 1 of Bloembergen[5] for further details.

The value of the wave vector mismatch Δk is very important. Only for $\Delta k = 0$ will large secular variations of the amplitudes and phases occur. Otherwise oscillations of small amplitude are the rule. In that case the parametric approximation suffices, where one or more fields are considered as constant parameters. Since momentum or wave vector matching occurs usually only for a few well defined sets of waves in a dispersive medium, it is possible to focus our attention on a small number of electromagnetic modes.[5]

5. Polarization Cubic in the Electric Field Amplitudes

In media with inversion symmetry this is always the dominant nonlinearity.[12] It is described by a fourth rank nonlinear susceptibility tensor whose elements may be calculated by reiterating the perturbation procedure outlined in Section 3. The ith component of the polarization at a combination frequency $\omega_4 = -\omega_1+\omega_2+\omega_3$ is given by

$$P_i(\omega_4) = \chi^{NL}_{ijkl}(-\omega_4,-\omega_1,\omega_2,\omega_3)\, E_j^*(\omega_1)\, E_k(\omega_2)\, E_l(\omega_3).$$

This nonlinearity will, in general, provide a coupling between four electromagnetic waves. If the polarization vectors are $\hat{a}_1$, $\hat{a}_2$, $\hat{a}_3$ and $\hat{a}_4$ respectively and we write

$$\chi^{NL}_{1234} = \hat{a}_1^*\,\hat{a}_2 : \chi(-\omega_1,\omega_2,\omega_3,-\omega_4) : \hat{a}_3\hat{a}_4^*$$

$$\chi^{NL}_{ij} = \hat{a}_i^*\,\hat{a}_i : \chi(-\omega_i,\omega_i,+\omega_j,-\omega_j) : \hat{a}_j\hat{a}_j^*$$

and the wave vector mismatch $\Delta k\hat{z} = \mathbf{k}_2+\mathbf{k}_3-\mathbf{k}_1-\mathbf{k}_4$ is introduced, the four coupled amplitude equations take the form

$$\frac{\partial A_1}{\partial z} + \frac{1}{v_{g_1}}\frac{\partial A_1}{\partial t} = +2\pi i(\omega_1/n_1c)\,[\chi^{NL}_{1234}\, A_2\, A_3\, A_4^*\exp(i\Delta kz) + \sum_{j=1}^{4}\chi^{NL}_{1j}\, A_1\, A_j\, A_j^*] \qquad (20a)$$

$$\frac{\partial A_2^*}{\partial z} + \frac{1}{v_{g_2}} \frac{\partial A_2^*}{\partial t} = -2\pi i(\omega_2/n_2\, c)\,[\chi^{NL}_{1234}\, A_1^*\, A_3\, A_4^* \exp(i\Delta kz) + \sum_{j=1}^{4} \chi^{NL}_{2j}\, A_2^*\, A_j\, A_j^*] \quad (20b)$$

$$\frac{\partial A_3}{\partial z} + \frac{1}{v_{g_3}} \frac{\partial A_3}{\partial t} = +2\pi i(\omega_3/n_3\, c)\,[\chi^{NL}_{1234}\, A_1\, A_2^*\, A_4 \exp(-i\Delta kz) + \sum_{j=1}^{4} \chi^{NL}_{3j}\, A_3\, A_j\, A_j^*] \quad (20c)$$

$$\frac{\partial A_4^*}{\partial z} + \frac{1}{v_{g_4}} \frac{\partial A_4^*}{\partial t} = -2\pi i(\omega_4/n_4\, c)\,[\chi^{NL}_{1234}\, A_1\, A_2^*\, A_3^* \exp(-i\Delta kz) + \sum_{j=1}^{4} \chi^{NL}_{4j}\, A_4^*\, A_j\, A_j^*]. \quad (20d)$$

Depending on whether the fourth rank susceptibility tensor element is real or imaginary, and on whether some of the frequencies are identical or different, a large variety of physical phenomena can be accounted for. Here the more important cases are listed, while Minck *et al.*[13] and Bloembergen[14] give a more detailed review.

(1) Third harmonic generation; $-\omega_1 = \omega_2 = \omega_3\ (\equiv\omega)$:

$$P^{NL}(3\omega) = \chi(-3\omega, \omega, \omega, \omega)\, A^3 \exp(3i\mathbf{k}\,.\,\mathbf{r} - 3i\omega t).$$

This polarization will radiate at the frequency 3ω.

(2) Two photon absorption: $\omega_1 = \omega_2 = \omega_3$

$$P^{NL}(\omega) = i\,\chi''_{NL}(-\omega, -\omega, \omega, +\omega)\, A|A|^2 \exp(i\mathbf{k}\,.\,\mathbf{r} - i\omega t).$$

The nonlinear susceptibility is positive pure imaginary, and has resonance at $2\omega = \omega_{ab}$, where a and b represent two energy levels with the same parity. The absorption becomes strong at high intensities.

(3) Saturable absorption; $+\omega_1 = +\omega_2 = \omega_3$: The same expression as under (2) applies, but now χ_{NL} is negative and has a resonance for $\omega = \omega_{ab}$. The absorption becomes weak at high intensity. The effect is used in bleachable filters.

(4) Intensity dependent index of refraction; $+\omega_1 = \omega_2 = \omega_3 = \omega_4$:

$$P^{NL}(\omega) = \chi'(-\omega, -\omega, \omega, +\omega)\, A|A|^2 \exp(i\mathbf{k}\,.\,\mathbf{r} - i\omega t).$$

(5) Stimulated Raman effect; $-\omega_1 = +\omega_4 = \omega_s$ and $\omega_2 = -\omega_3 = \omega_L$:

$$P^{NL}(\omega_s) = i\chi''(-\omega_s, \omega_s, \omega_L, -\omega_L)\, A_s|A_L|^2 \exp(i\mathbf{k}_s\,.\,\mathbf{r} - i\omega_s t).$$

In this case the nonlinear susceptibility is pure imaginary and negative, while the difference between the laser and stokes frequency is at a resonance of the material system, $\omega_L - \omega_s = \omega_{ab}$.

(6) Antistokes generation; $\omega_2 = \omega_3 = \omega_L$, $\omega_4 = \omega_L + \omega_{ab} = \omega_{as}$, $\omega_1 = \omega_L - \omega_{ab}$:

$$P^{NL}(\omega_{as}) = \chi^{NL}(-\omega_{as}, \omega_L, \omega_L, -\omega_s) A_L{}^2 A_s{}^* \times \exp\{i(2\mathbf{k}_L - \mathbf{k}_s) \,.\, \mathbf{r} - i(2\omega_L - \omega_s)t\}$$

The antistokes radiation will only be intense in the immediate vicinity of the momentum matching direction, where $\mathbf{k}_{as} = 2\mathbf{k}_L - \mathbf{k}_s$.

(7) Stimulated Rayleigh scattering; $+\omega_4 = -\omega_1 = \omega_L'$ and $-\omega_3 = \omega_2 = \omega_L$:

Formally this process is the same as the stimulated Raman scattering from a rotational level. The stokes frequency ω_L' is now a frequency slightly smaller than the laser frequency, so that $\omega_L - \omega_L' = \tau_c{}^{-1}$, where τ_c is the relaxation time for molecular reorientation.

(8) Modulation of the index of refraction:

$$P^{NL}(\omega_3 + \omega_L - \omega_L') = \chi(-\omega_3 - \omega_L + \omega_L', \omega_3, \omega_L, -\omega_L') A_L A_L'{}^* A_3 \times \exp\{i(\mathbf{k}_L - \mathbf{k}_L' + \mathbf{k}_3) \,.\, \mathbf{r} - i(\omega_3 + \omega_L - \omega_L')t\}.$$

In this case all four frequencies are different, but ω_L and ω_L' are close together, so that $\omega_L - \omega_L' \leqslant \tau_c{}^{-1}$. Side bands are created around the frequency ω_3. The susceptibility is in general complex.

(9) Self-focussing of light:

In this case the frequencies are all equal as for the intensity dependent index of refraction. The two wave vectors $\mathbf{k}_3$ and $\mathbf{k}_4$ make a small angle with the wave vector $\mathbf{k}_2 = \mathbf{k}_1 = \mathbf{k}_L$. Since the wave vectors have equal length, there is a slight mismatch $\Delta\mathbf{k} = 2\mathbf{k}_L - \mathbf{k}_3 - \mathbf{k}_4$.

The macroscopic symmetry properties of these effects may be discussed along the same lines as indicated in Section 3 for the third rank susceptibility tensor. Only a few illustrations will be mentioned here.

Third harmonic generation in an isotropic fluid is described by one scalar element according to the vector equation

$$\mathbf{P}(3\omega) = C(\mathbf{E} \,.\, \mathbf{E})\,\mathbf{E},$$

where $C = \chi_{xxxx} = \chi_{yyyy} = \chi_{zzzz} = 3\chi_{xxyy}$ etc. The polarization for the third harmonic is parallel to the linearly polarized fundamental field. Circular polarization yields no third harmonic. In a cubic crystal two independent

constants (χ_{xxxx} and χ_{xxyy}) are necessary to describe the tensor. For propagation along a cubic axis circularly polarized light will give a third harmonic polarized in the opposite sense.

The second harmonic generation induced in an isotropic fluid by the application of a dc electric field is described by two independent constants

$$\mathbf{P}^{NL}(2\omega) = C_1 (\mathbf{E}(\omega) \cdot \mathbf{E}(\omega)) \mathbf{E}_{dc} + C_2 (\mathbf{E}(\omega) \cdot \mathbf{E}_{dc}) \mathbf{E}(\omega).$$

If the dispersion is negligible and the Kleinman symmetry relation is valid, then $C_1 = C_2$ and only one independent constant remains. The three constants introduced by Mayer,[15] who measured this effect as a function of the relative directions of polarization of the incident beam and the dc electric field, should have a well defined ratio, consistent with the relation, in our notation,

$$\chi_{xxxx}(-2\omega, 0, \omega, \omega) : \chi_{xxyy}(-2\omega, 0, \omega, \omega) : \chi_{xyxy}(-2\omega, 0, \omega, \omega) = 3 : 1 : 2.$$

This has been confirmed experimentally.

The experimentally observed magnitude of nonlinear susceptibilities may be compared with the theoretical calculation outlined in Section 2. As eqn (14) shows, the matrix elements between all excited states must be known. If the denominators are all taken to be equal, (this is a good approximation if the light frequencies are small compared to any average excitation frequency Ω), a closure rule may be applied and the susceptibilities may be expressed in terms of moments of ground state wave functions alone. In this approximation there is no dispersion, and one finds for the susceptibilities of bound electrons in the ground state $|g\rangle$, the following expressions, where

$$\tilde{x}_i = x_i - \langle g|x_i|g\rangle,$$

$$\chi_{ij}^{(1)} = \frac{2Ne^2}{\hbar\Omega} \langle g|\tilde{x}_i \tilde{x}_j|g\rangle$$

$$\chi_{ijk}^{NL} = -\frac{3Ne^3}{\hbar^2\Omega^2} \langle g|\hat{x}_i \hat{x}_j \hat{x}_k|g\rangle$$

$$\chi_{ijkl}^{NL} = \frac{4Ne^4}{\hbar^3\Omega^3} \langle g|\tilde{x}_i \tilde{x}_j \tilde{x}_k \tilde{x}_l|g\rangle - 2\langle g|\tilde{x}_i \tilde{x}_j|g\rangle\langle g|\tilde{x}_k \tilde{x}_l|g\rangle.$$

We assume N equivalent valence electronic orbitals per unit volume. For a given crystal structure one has an additional geometrical factor to take account of the orientations of the orbitals in the unit cell. The observed linear susceptibility may be used to determine the effective mean excitation frequency Ω. The nonlinear susceptibilities have thus been calculated for the simple crystal structures of the III–V and group IV semiconductors, on the basis of various models for the ground state wave function.

Table I compares the nonlinear susceptibility describing second harmonic generation of the 10·6 μ CO_2 laser line with theory, and Table II compares the mixing of the 10·6 μ and 9·6 μ light of two CO_2 laser lines to yield a new combination frequency $2\omega_1-\omega_2$ of wavelength 11·6 μ, in these materials. The agreement is satisfactory if a suitable model for the ground state wave function is chosen. The theory of the nonlinear susceptibilities is plagued by the same difficulties as the theory for the linear susceptibilities. Since the lowest order susceptibility depends on the lack of parity of the wave function, it is quite sensitive to the amount of asymmetry ,which is rather well described by Phillips model of electronegativity.

TABLE I

Comparison between experimental and theoretical values of $2\chi_{14}(-2\omega, \omega, \omega)$ in units of 10^{-6}e.s.u. The harmonic generation is due to valence band electrons, while ω corresponds to the 10·6 μ CO_2 laser line.

Crystal	InAs	GaAs	GaP	GaSb
Experiment[a]	+1·0	+0·45	+0·26	+1·51
Theory[b]	−1·5	−0·43	−0·21	−7·8
Theory[c]	+0·96	+0·57	+0·34	+1·2
Theory[d]	+2·05	+0·95	+0·60	+0·80
Theory[e]	+1·1	+0·85	+0·2	+1·8

[a] Wynne, J. J. Ph.D. Thesis, Harvard University (1969); Bloembergen, N. and Wynne, J. J. *Phys. Rev.* **188,** 1211 (1969).
[b] Jha, S. S. and Bloembergen, N. *Phys. Rev.* **171,** 891 (1968).
[c] Levine, B. F. *Phys. Rev. Lett.* **22,** 787 (1969).
[d] Flytzanis, C. and Ducuing, J. *Phys. Rev.* **178,** 1218 (1969).
[e] Flytzanis, C. *C.r. hebd. Seanc. Acad. Sci., Paris.* **B267,** 555 (1968).

TABLE II

Comparison between experimental and theoretical values of the nonlinear mixing susceptibility $4\chi^{NL}_{\xi\xi\xi\xi}(-2\omega_1+\omega_2, \omega_1, \omega_1, -\omega_2)$ in units of 10^{-10}e.s.u. The mixing is due to valence band electrons, ω_1 and ω_2 correspond to the 10·6 μ and 9·6 μ CO_2 laser lines respectively.

Crystal	Ge	Si	GaAs
Experiment[a]	+1·5	0·08	+0·10
Theory[b]	−0·88	−0·063	−0·13
Theory[c]	−1·6	−0·53	—

[a] Wynne, J. J. *Phys. Rev.* **178,** 1295 (1969).
[b] Jha, S. S. and Bloembergen, N. *Phys. Rev.* **171,** 891 (1968).
[c] Levine, B. F. *Phys. Rev. Lett.* **22,** 787 (1969). (Since references [a] and [c] use a different definition of the complex amplitudes, the value 4χ has been tabulated, where χ follows the definition used in this paper).

6. Stimulated Raman Effect[14]

In the stimulated Raman effect two waves are incident on the material system, one at the laser frequency ω_L and one at the stokes frequency ω_s, so that $\omega_L - \omega_s$ corresponds to a resonant frequency $\omega_{ba} = \omega_v$ of the material. This is often a molecular vibrational frequency, but it may correspond to an electronic transition. Quanta are scattered from the laser beam to the stokes beam, while the material makes a transition from state $|a\rangle$ to $|b\rangle$. This process is described by a Raman susceptibility, which is negative imaginary at the resonance condition $\omega_L - \omega_s = \omega_v$. The amplitude of the stokes wave obeys eqn (20a), which in this case for a process stationary in time reduces to

$$\frac{dA_s}{dz} = +2\pi(\omega_s/n_s c)\,(-\chi''_{\text{Raman}})\,|A_L|^2 A_s. \tag{21}$$

The amplitude grows exponentially, as the nonlinear Raman polarization is always leading the electric field A_s by 90° and continues to do work on the wave. At the same time the laser beam would be attenuated because we have the relationship

$$\chi_{\text{Raman}} = +\chi_{jjii}(-\omega_s, \omega_s, -\omega_L, \omega_L) = \chi^*_{iijj}(-\omega_L, \omega_L, -\omega_s, \omega_s).$$

Thus eqn (20b) becomes

$$\frac{dA_L}{dz} = -2\pi(\omega_L/n_L c)\,(-\chi''_{\text{Raman}})\,|A_s|^2 A_L. \tag{22}$$

This equation describes the depletion of the laser beam. Initially the laser intensity $|A_L|^2$ may be taken as a constant parameter. The stokes intensity then grows exponentially.

$$\mathcal{I}_s = \mathcal{I}_0 \exp(g_s z) \tag{23}$$

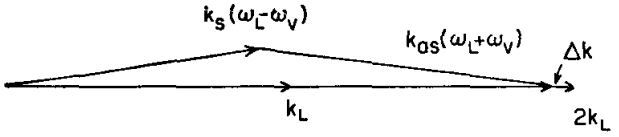
with a Raman gain coefficient given by

$$g_s = 4\pi(\omega_s/n_s c)\,|\chi''_{\text{Raman}}|\,|A_L|^2. \tag{24}$$

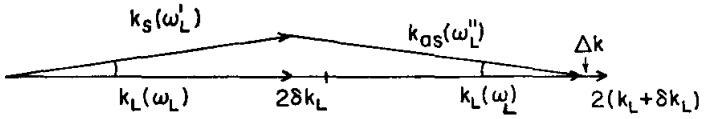
This gain constant is also directly related to the cross section for spontaneous Raman scattering, because the same material matrix elements occur in the calculation of the nonlinear susceptibility describing the stimulated counterpart of the spontaneous emission.

When an antistokes frequency $\omega_{as} = \omega_L + \omega_v$ and a laser frequency ω_L are considered together, the exponential loss will occur at ω_{as}, while ω_L will exhibit exponential gain. When the population of the two vibrational levels is inverted, the nonlinear susceptibility changes sign and gains and losses

would be interchanged. With a non-inverted population it is nevertheless possible to have exponential gain at the antistokes frequency, but only in particular directions where there exists a close coupling between stokes and antistokes waves, i.e. for small $\Delta k\hat{z} = \mathbf{k}_s + \mathbf{k}_{as} - 2\mathbf{k}_L$. Due to the normal colour



(a)



(b)

Fig. 1a. The wave vector relationship in stimulated Raman scattering of stokes and anti-stokes radiation.

Fig. 1b. The wave vector relationship of scattering of light without significant change of frequency due to the intensity dependent index of refraction. The laser pump wave vector is corrected by $\delta k_L = \frac{1}{2}k_L\theta^2_{\text{opt}}$

dispersion in most liquids the antistokes gain occurs in the cone with an apex angle of a few degrees, as shown in Fig. 1a. If the laser field is considered as a constant parameter, eqns (20a and d) with $A_1 = A_s$ and $A_4 = A_{as}$ become in the steady state regime

$$\frac{dA_s}{dz} = +2\pi i(\omega_s/n_s\, c)\,\chi^{NL}_{sL}\,|A_L|^2 A_s + 2\pi i(\omega_s/n_s\, c)\,\chi_{saLL}\,A_L^2\,A_{as}^*\exp(i\Delta kz) \quad (25a)$$

$$\frac{dA_{as}^*}{dz} = -2\pi i(\omega_{as}/n_{as}\, c)\,\chi_{asLL}\,A_L^{*2}A_s\exp(-i\Delta kz) - 2\pi i(\omega_{as}/n_{as}\, c) \times \chi^{NL}_{aL}\,|A_L|^2\,A_{as}^*. \quad (25b)$$

This is a set of two linear coupled equations in A_s and A_{as}^* and can be solved exactly, by anticipating exponential solutions of the form $A_s\exp(ik_sz + i\Delta Kz)$ and $A_{as}^*\exp(-ik_{as}z + i\Delta Kz)$. Substitution into eqns (25) leads to a quadratic equation in ΔK and the intensity gain coefficient is 2 Im ΔK or,

$$g_s = \pm\, 2\,\text{Im}[\tfrac{1}{4}(\Delta k)^2 + 2\pi(\omega_s/n_s\, c)\,\chi^{NL}|A_L|^2\Delta k]^{1/2}. \quad (26)$$

Here we have made the (unnecessary) approximation that the dispersion in the nonlinear coefficients can be ignored. For large momentum mismatch, $|\Delta k| > [2\pi(\omega_s/n_s c)|\chi^{NL}|]^{1/2}|A_L|$, the square root may be expanded and g_s reduces to eqn (24). The stokes and antistokes waves are uncoupled and have the single wave gain or loss. Near $\Delta k = 0$ the normal modes are mixtures of stokes and antistokes. A wave with 70% stokes character and 30% antistokes character will have an exponential gain, while the mode with 70% antistokes character in the same direction will decay exponentially. One thus observes an exponential gain for antistokes light because it is dragged along by the dominant stokes character of the mode. Note that for exact momentum matching, $\Delta k = 0$, there is neither gain nor loss. The normal modes have exactly half stokes and antistokes character.

Many further examples of this coupled mode behaviour will now be given. In fact, the pure stokes generation may be considered as the parametric down conversion of a laser wave into the stokes wave and a vibrational wave. In this description the response of the material represented by the off diagonal element of the density matrix ρ_{ab} is explicitly displayed, whereas in eqn (21) it is hidden in the Raman susceptibility. The dispersive behaviour of the optical phonon wave is described by

$$\omega_v^2 = \omega_{v,0}^2 + \beta k_v^2,$$

where $\omega_{v,0}$ is the vibrational frequency of an isolated molecule. The optical phonon branch is usually very narrow and the dispersion constant β is correspondingly small, so that the stokes shift is essentially the same in all directions. A damping time of the optical phonon wave τ_v is introduced phenomenologically. It represents the effect of collisions with acoustical phonons or other degrees of freedom. The width of the spontaneously emitted Raman line is proportional to τ_v^{-1}.

The coupling of the light waves and the vibrations may be described in terms of a classical model, in which the polarizability of the molecule is a linear function of the vibrational coordinate (internuclear distance R),

$$\alpha = \alpha_0 + (\partial\alpha/\partial Q_v)\, Q_v,$$

where

$$Q_v = R\,(2\rho)^{1/2} \exp(-i\mathbf{k}_v \cdot \mathbf{r}) \tag{27}$$

is the optical phonon coordinate, and ρ is the reduced vibrational mass density. From the coupling term in the time-averaged potential energy,

$$U_{\text{int}} = -(\partial\alpha/\partial Q_v)\, Q_v\, E_L^* E_s + \text{complex conjugate}, \tag{28}$$

the following set of coupled equations may be derived for Q_v^* and E_s,

$$\ddot{Q}_v^* - \beta|k_L - k_s|^2 Q_v^* + \omega_{v,0}{}^2 Q_v^* + (2i/\tau_v)\dot{Q}_v^* = N(\partial\alpha/\partial Q_v) E_L^* E_s \quad (29)$$
$$\ddot{E}_s + \omega_s{}^2 E_s + (2/i\tau_s)\dot{E}_s = 4\pi\omega_s{}^2 N(\partial\alpha/\partial Q_v)^* E_L Q_v^*,$$

where the dots denote time differentiation. Phenomenological damping terms have been introduced for the vibrational and the stokes wave. Since the coupling term is small, the set of coupled wave equations may be reduced to a set of coupled amplitude equations

$$\dot{A}_v^* + (A_v^*/\tau_v) = (\omega_v/2i) N(\partial\alpha/\partial Q) A_L^* A_s$$
$$\dot{A}_s + (A_s/\tau_s) = (+2\pi i\omega_s) N(\partial\alpha/\partial Q)^* A_L A_v^* \quad (30)$$

In these equations the amplitudes are represented as growing in time. The corresponding problem of spatially growing waves is obtained by replacing $\partial/\partial t$ by $v_{\text{group}}(\partial/\partial z)$. The solution of this problem of two coupled waves is already familiar. There is one coupled mode with exponential gain and another mode with exponential loss. Since the optical phonon is heavily damped, the mode with exponential gain has predominantly stokes light character. To the extent that $\dot{A}_v^*$ or $v_{\text{group}}\,\partial A_v^*/\partial z$ may be ignored compared to the damping term $A_v^*\,\tau_v{}^{-1}$, the steady state value

$$A_v^* = (\tau_v\omega_v/2i) N(\partial\alpha/\partial Q) A_L^* A_s \quad (31)$$

may be substituted directly into the equation for A_s. A stokes wave with an exponentially growing amplitude results, if

$$\pi(\omega_s\omega_v) N^2(\partial\alpha/\partial Q)^2 |A_L|^2 \tau_v \geqslant \tau_s{}^{-1}. \quad (32)$$

The term on the left is the Raman gain, the term on the right represents the amplitude decay constant of the stokes mode in the stokes resonator, caused by reflectivity and scattering losses. The gain in time or in space may be equated to the expression for the Raman gain g_s in terms of the Raman susceptibility or the spontaneous Raman cross section. One thus finds for the Raman susceptibility, in terms of this classical model for the Raman effect

$$\chi''_{\text{Raman}} = -n_s{}^2 N^2(\partial\alpha/\partial Q)^2\tau_v/2\omega_v. \quad (33)$$

Antistokes generation can be discussed in terms of this model by considering the following four waves, the laser wave, the optical phonon wave, the stokes wave and the antistokes wave, satisfying the following energy and momentum conservation rules

$$\omega_v = \omega_L - \omega_s = \omega_{as} - \omega_L,$$
$$\mathbf{k}_v = \mathbf{k}_L - \mathbf{k}_s = \mathbf{k}_{as} - \mathbf{k}_L.$$

The laser wave is eliminated in the parametric approximation by considering the laser field as a fixed parameter. The optical phonon wave may be eliminated from the coupled equations, because its group velocity is near zero and it is heavily damped. In this manner one obtains two equations for the coupling between stokes and antistokes which leads again to a gain constant, identical to eqn (26).

The two formulations are therefore equivalent, as far as the stimulated Raman effect is concerned. It should be remembered that the Raman effect is not always associated with a vibrational excitation. It could be an electronic excitation in which case the optical phonon should be replaced by an exciton wave. Or it could be rotational excitation, in which case a "rotational wave" would be involved, etc. The stimulated Brillouin and Rayleigh scattering may be considered in a similar manner. The interaction between the light waves and any other coherent excitation or boson wave can thus be described, and seemingly very different physical phenomena can be discussed from a common point of view.

7. Inelastic Rayleigh Scattering from Anisotropic Molecules[14]

In fluids with optically anisotropic molecules the dominant nonlinear effect is often caused by the reorientation of the molecules subjected to the strong electric fields of one or more laser beams. Consider an axially symmetric molecule with polarizabilities $\alpha_{\|}$ and $\alpha_{\perp}$, subjected to two parallel fields $E_L(\omega_L)$ and $E_L'(\omega_L')$, which make an angle θ with the axis of symmetry.

The orientation dependent part of the dielectric energy may be written as

$$U = \tfrac{1}{6}(\alpha_{\|}-\alpha_{\perp})\,(1-3\cos^2\theta)\,[A_L A_L'^{*}\exp(-i\Delta\omega t) + A_L^{*}A_L'\exp(+i\Delta\omega t) + |A_L|^2+|A_L'|^2]. \tag{34}$$

Here $\Delta\omega = \omega_L-\omega_L'$ and the rapidly varying term at $\omega_L+\omega_L'$ has been ignored. There is a corresponding torque $\tau' = -\partial U/\partial\theta$ acting on the molecule. In addition there is the viscous torque responsible for a rotational Brownian motion. The Fokker–Planck equation for this motion was given by Debye

$$\frac{\partial f}{\partial t} = \frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left[\frac{kT}{\zeta}\sin\theta\frac{\partial f}{\partial\theta} - \frac{\tau' f}{\zeta}\right]. \tag{35}$$

For a sphere of radius a in a fluid with viscosity η the damping constant $\zeta = 8\pi\eta a^3$. To lowest order in U/kT the steady state solution for the rotation distribution function is, if we introduce the orientation relaxation time

$\tau = \zeta/6kT$,

$$f = \frac{1}{4\pi} + \frac{1}{12\pi kT}(\alpha_{||} - \alpha_{\perp})(3\cos^2\theta - 1)\left[|A_L|^2 + |A_L'|^2 + \frac{A_L A_L'^*}{1 - i\Delta\omega\tau}\right.$$

$$\left. \times \exp(-i\Delta\omega\tau) + \frac{A_L^* A_L'}{1 + i\Delta\omega\tau}\exp(+i\Delta\omega\tau)\right]. \qquad (36)$$

From this nonisotropic distribution, a polarization cubic in the electric field strength may be calculated,

$$P^{NL} = \tfrac{1}{3}N_0|\alpha_{||} - \alpha_{\perp}|\int_0^{\pi}(3\cos^2\theta - 1) f 2\pi \sin\theta \, d\theta [A_L \exp(-i\omega_L t)$$

$$+ A_L^* \exp(+i\omega_L t) + A_L' \exp(-i\omega_L' t) + A_L'^* \exp(+i\omega_L' t)]. \qquad (37)$$

This polarization describes not only the intensity dependent index of refraction, but also the modulation of this index at the beat frequency. This provides a coupling between waves spaced in frequency by $\Delta\omega$. Due to the phase shifts when $\Delta\omega\tau$ is comparable or larger than unity, there will be a polarization at ω_L', proportional to the intensity $|A_L|^2$, and leading the electric field A_L' in phase. This will provide exponential growth at ω_L' of this wave, the maximum gain occurring for $\omega_L' = \omega_L - \tau^{-1}$. This is the stimulated inelastic Rayleigh scattering.

It is instructive to consider also the coupling with the antistokes Rayleigh component and to allow the two waves to make a small angle θ with the direction of the strong laser beam E_L. We have again the set of coupled equations (25a) and (25b), but the nonlinear susceptibility constants are, for the orientational Rayleigh case,

$$\chi_{sL} = \chi_{aL}^* = \frac{4N_0(\alpha_{||} - \alpha_{\perp})^2}{45kT}\left(1 + \frac{1}{1 + i\Delta\omega\tau}\right) \qquad (38a)$$

$$\chi_{saLL} = \chi^*_{asLL} = \frac{4N_0(\alpha_{||} - \alpha_{\perp})^2}{45kT}\frac{1}{1 + i\Delta\omega\tau}. \qquad (38b)$$

The wave vector mismatch Δk may be determined from the diagram in Fig. 1b. It should be kept in mind that the wave vector of the laser beam itself is changed, by the intensity dependent index, from the original value $k_L = \omega_L c^{-1}\varepsilon_0^{1/2}$ to the value $k_L(1 + \frac{1}{2}\theta^2_{\text{opt}})$.

Here a short hand notation is introduced, the meaning of which will shortly become apparent,

$$\theta^2_{\text{opt}} = \frac{N_0(\alpha_{||} - \alpha_{\perp})^2 |A_L|^2 16\pi}{45kT\varepsilon_0}$$

The momentum mismatch may then be expressed in the form, for small angles θ,

$$\Delta k = k_L(\theta^2 + \theta^2{}_{\text{opt}}).$$

Solving the two by two determinantal problem for the two coupled amplitude equations, one finds for the intensity gain constant

$$g = \frac{\theta k_L}{\sqrt{2}} \left\{ \left[\theta^4 + \frac{4\theta^2{}_{\text{opt}}}{1+\Delta\omega^2\tau^2}(\theta^2{}_{\text{opt}} - \theta^2) \right]^{\frac{1}{2}} + \frac{2\theta^2{}_{\text{opt}}}{1+\Delta\omega^2\tau^2} - \theta^2 \right\}^{\frac{1}{2}}. \quad (39)$$

This gain is a maximum for $\theta = \theta_{\text{opt}}$, and its value is then

$$g_{\max} = \frac{k_L\theta^2{}_{\text{opt}}}{1+\Delta\omega^2\tau^2}. \quad (40)$$

The behaviour is plotted in Fig. 2. For $\theta \gg \theta_{\text{opt}}$ the gain is independent of θ.

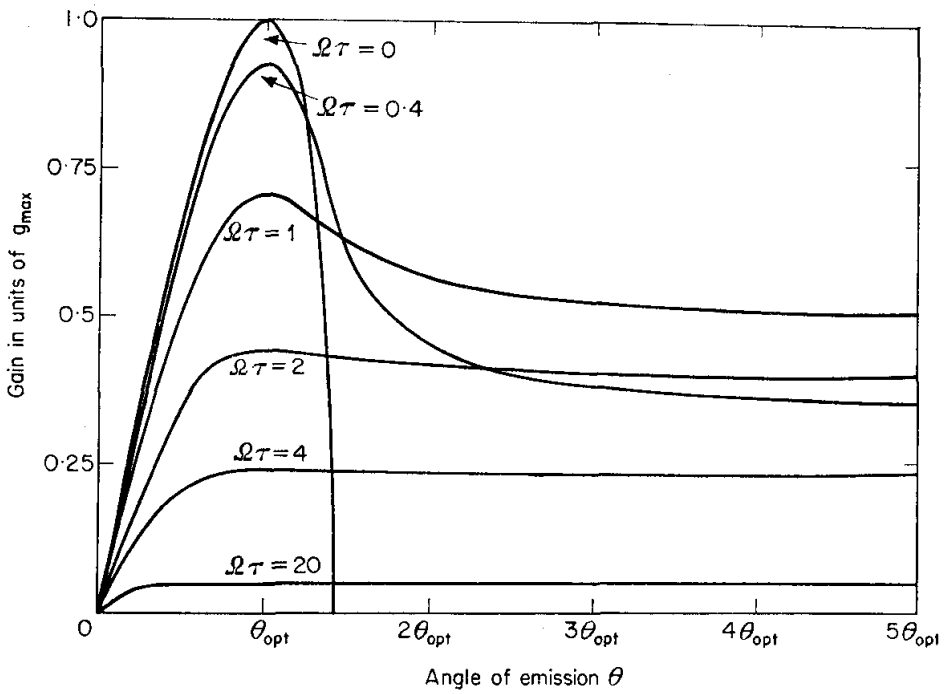


Fig. 2. The dependence of the gain constant on angle and small frequency shift $\Omega = \omega_L - \omega_L' = \omega_L'' - \omega_L$ for the near degenerate scattering process shown in figure 1b (after Chiao, Kelley and Garmire, *Phys. Rev. Lett.* **17**, 1158, (1966)).

There we have the uncoupled behaviour and the stokes gain is maximum for $\Delta\omega\tau = 1$ and vanishes for $\Delta\omega = 0$. This stimulated inelastic Rayleigh scattering has been observed in the backward direction.

For small angles θ there is gain at $\Delta\omega = 0$ for $0 < \theta < \sqrt{2}\,\theta_{\text{opt}}$. In this case two waves grow at the same frequency, but slightly off-axis from the original

laser beam. The growth of these off-axis components signifies the onset of self-focussing. Since the index of refraction is highest on the centre axis of the laser beam, where the intensity is highest, light rays will be bent towards the axis, since in the geometrical optics approximation the curvature of the ray is proportional to the transverse index gradient. The laser beam orientates the molecules in the fluid in such a manner as to create a converging lens. Since so many pairs of off-axis waves are involved, it is better to take the transverse variation of the field amplitudes and the concomitant diffraction effects into account by the following wave equation

$$\frac{\partial^2 A}{\partial x^2} + \frac{\partial^2 A}{\partial y^2} + 2ik_z \frac{\partial A}{\partial z} = -4\pi k_z{}^2 \chi^{NL} |A|^2 A. \tag{41}$$

We shall not discuss the solution of this equation, but it does lead to self focussing and possibly the creation of bright filaments ranging in diameter from 5–50 microns. In these filaments the intensity is much higher than the nominal flux density in the incident laser beam. This self-focussing phenomenon explains many anomalies in the observation of other nonlinear effects, especially the anomalously high Raman gain. The observed frequency broadening of stokes and antistokes components is caused by the modulation of the index of refraction. It is described by eqns (36 and 37), which give polarization components at new frequencies $2\omega_L - \omega_L'$, etc. Both effects are especially pronounced in fluids with optically anisotropic molecules.

8. Stimulated Brillouin Effect

In this case the role of the optical phonon is taken over by an acoustical phonon. The main distinction between the stimulated Raman effect and the stimulated Brillouin effect lies in the different dispersion characteristics of the acoustical and optical phonon branches. The Brillouin frequency shift depends on the angle of scattering θ. This dependence can be derived from the resonant conditions of energy and momentum conservation, viz;

$$\mathbf{k}_{ac} = \mathbf{k}_L - \mathbf{k}_s \quad \text{and} \quad \omega_{ac} = v_{ac}|k_{ac}|\gamma^{-\frac{1}{2}} = \omega_L - \omega_s.$$

To a very good approximation the Brillouin frequency shift in isotropic media is given by,

$$\omega_B = 2\omega_L(nv_{ac}/c\gamma^{\frac{1}{2}}) \sin(\theta/2), \tag{42}$$

where $\gamma = C_p/C_v$ the ratio of the specific heats at constant pressure and density respectively, and v_{ac} is the low frequency limit of the acoustic velocity.

Again we have a coupled wave problem between one or more light waves and the acoustic wave. We write down the equations for an isotropic fluid,

where the density fluctuations

$$\rho = \rho_0 + \rho_1 \exp(i\mathbf{k}_{ac} \cdot \mathbf{r} - i\omega_{ac} t)$$

are coupled to a Brillouin stokes component travelling in the backward direction. In the near forward direction the frequency shift becomes very small and there would also be simultaneous coupling to an antistokes component.

The acoustic wave equation has the form

$$\frac{\partial^2 \rho_1^*}{\partial t^2} - \frac{v_{ac}^2}{\gamma} \nabla^2 \rho_1^* + \frac{2}{\tau_{ac}} \frac{\partial \rho_1^*}{\partial t} = \frac{1}{4\pi} \frac{\partial \varepsilon}{\partial \rho} \nabla^2 (E_s E_L^*), \quad (43)$$

where τ_{ac} is the damping time of the acoustic wave. The right-hand side represents the electrostrictive effect which drives the acoustic wave at the difference frequency between the Brillouin light wave and the laser wave.

The Maxwell equation for the Brillouin stokes component is

$$\nabla^2 E_s - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} [\varepsilon(\omega_s) E_s] = \frac{1}{c^2} \frac{\partial \varepsilon}{\partial \rho} \frac{\partial^2}{\partial t^2} (E_L \rho_1^*). \quad (44)$$

The right-hand side represents the change in polarization due to the photoelastic effect. For a medium obeying the Lorentz–Lorenz relation, $\partial\varepsilon/\partial\rho = (\varepsilon-1)(\varepsilon+2)/3\rho_0$.

The two coupled wave equations may now be treated in a manner similar to the treatment of the stimulated Raman effect. They can be reduced to two coupled amplitude equations. For a quasi steady-state situation, where only spatial growth has to be considered, the problem is again reduced to a two by two determinantal problem. Since the acoustic propagation velocity is small and the damping of the acoustic wave is large, we may again first solve for ρ_1 and substitute the steady-state value

$$\rho_1 = \frac{\tau_{ac}}{2i\omega_{ac}} \frac{1}{4\pi} \frac{\partial \varepsilon}{\partial \rho} |\mathbf{k}_L - \mathbf{k}_s|^2 A_s A_L^* \quad (45)$$

into the Brillouin wave equation. This leads to an exponential gain constant for the Brillouin wave. If we introduce the Brillouin shift

$$\omega_B^2 = (v_{ac}^2/\gamma) |\mathbf{k}_L - \mathbf{k}_s|^2,$$

the gain constant for the Brillouin stokes component may then be written in the form

$$g_B(\omega_s) = \frac{\omega_s}{n_s c} \rho_0 \left(\frac{\partial \varepsilon}{\partial \rho} \right)_T^2 |A_L|^2 \frac{\tau_{ac} |\mathbf{k}_L - \mathbf{k}_s|^2}{2(\omega_L - \omega_s)} [1 + \tau_{ac}^2 (\omega_L - \omega_s - \omega_B)^2]^{-1}. \quad (46)$$

This exponential gain has been demonstrated experimentally in a manner similar to that used to measure the gain constant in the stimulated Raman effect. The arrangement of a Brillouin backward stokes oscillator and a backward amplifier cell is shown schematically in Fig. 3a. By varying the

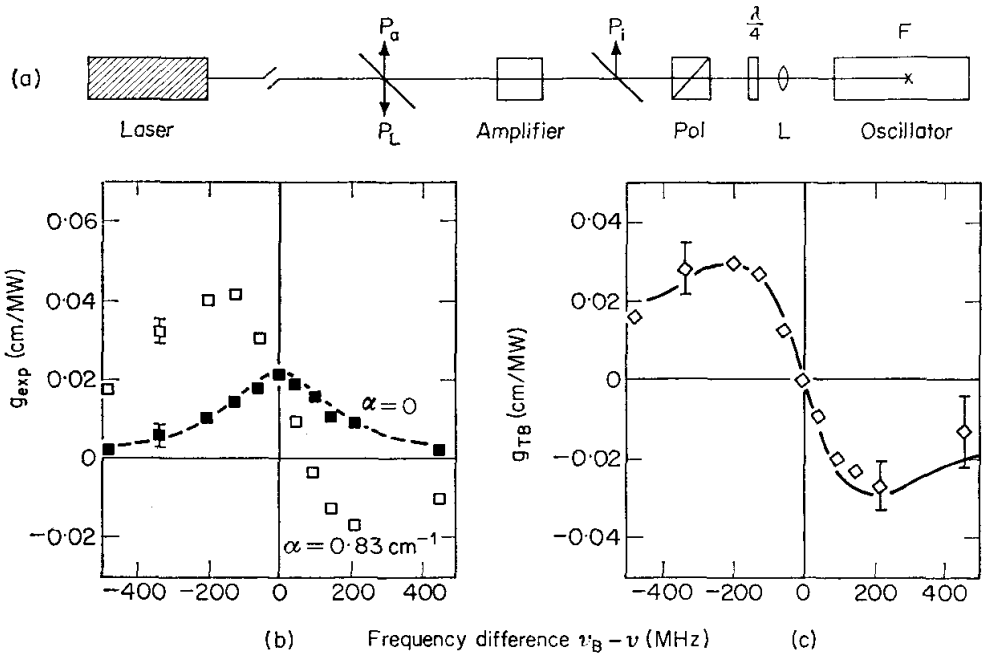


Fig. 3. (a) Diagram of the experimental arrangement to measure the backward Brillouin scattering gain. (b) The experimental data for a nonabsorbing liquid (solid squares) and the data with the addition of an absorbing material $-\alpha = 0{\cdot}83\ cm^{-1}$ (open squares). (c) Correction due to the thermally induced effect. g_{TB} is the difference between the experimental gain measured with and without absorption. The solid line is given by eqn (50). (After Pohl, Reinhold and Kaiser, *Phys. Rev. Lett.* **20**, 1141, (1968).)

composition of a liquid mixture in the oscillator cell, its frequency can be tuned through the gain profile of the liquid in the amplifier. The resulting curve is shown in Fig. 3b. When the liquid is absorbing, a very different result is obtained as shown by the open points. In this case the temperature changes and a new coupling to the dielectric constant must also be taken into account. It should be emphasized that these results are only valid in the quasi steady-state regime. For short laser pulses of duration comparable to or shorter than the acoustic damping time, transient effects must be taken into account, as discussed in the final section of this chapter.

9. Thermally Induced Brillouin and Rayleigh Scattering

In absorbing media the temperature variation and the concomitant changes in index of refraction must also be taken into account. Even in nonabsorbing

media there is an electrocaloric effect described by the intrinsic temperature dependence of the dielectric constant, $(\partial\varepsilon/\partial T)_\rho$. This term is, however, rather small compared with the effect due to $(\partial\varepsilon/\partial\rho)_T$ in combination with thermal expansion. The combined behaviour of the stokes and antistokes light waves, the density wave and the temperature wave must be considered. The temperature wave normally has a strictly diffusive behaviour but, in the case of second sound, a second time derivative of the temperature should be added to the equation. Unless one looks in the near forward direction, the two light waves are essentially uncoupled due to the momentum mismatch. We then have the following set of three coupled equations,

$$\nabla^2 E_s - \frac{\varepsilon_s}{c^2}\frac{\partial^2 E_s}{\partial t^2} = \left(\frac{\partial\varepsilon}{\partial T}\right)_\rho \frac{1}{c^2}\frac{\partial^2(E_L\theta_1^*)}{\partial t^2} + \left(\frac{\partial\varepsilon}{\partial\rho}\right)_T \frac{1}{c^2}\frac{\partial^2(E_L\rho_1^*)}{\partial t^2}, \tag{47a}$$

$$-\kappa\nabla^2\theta_1^* + \frac{\partial\theta_1^*}{\partial t} - \frac{(\gamma-1)}{\rho_0\beta}\frac{\partial\rho_1^*}{\partial t} = \frac{\alpha c n}{\rho_0 C_v 8\pi} E_L^* E_s + \frac{1}{4\pi} T_0 \left(\frac{\partial\varepsilon}{\partial T}\right)_\rho \times \frac{\partial}{\partial t}(E_L^* E_s), \tag{47b}$$

$$\frac{\partial^2\rho_1^*}{\partial t^2} - \frac{v_{ac}^2}{\gamma}\nabla^2\rho_1^* + \frac{2}{\tau_{ac}}\frac{\partial\rho_1^*}{\partial t} - \frac{\beta\rho_0 v_{ac}^2}{\gamma}\nabla^2\theta_1^* = \frac{1}{4\pi}\rho_0\left(\frac{\partial\varepsilon}{\partial\rho}\right)_T \times \nabla^2(E_L^* E_s). \tag{47c}$$

Here the temperature variations at a frequency close to $\omega_L - \omega_s$ are denoted by $\theta_1 = T - T_0$; $\kappa\,(= \lambda/\rho_0 C_v)$ is the thermal diffusivity, C_v is the specific heat at constant volume, α is the absorption coefficient for the light, β is the volume thermal expansion coefficient. The temperature variations are driven by adiabatic expansion, which can often be ignored in dense liquids ($\gamma \approx 1$), by light absorption and by the electrocaloric effect. We have ignored the absorption proportional to $\alpha(|E_L|^2 + |E_s|^2)$ which will give rise to a linearly increasing transient effect. The longitudinal compressive type acoustic wave equation is the same as in the preceding section, except for the addition of a term proportional to the thermal derivative. The light wave equation has an added term proportional to $(\partial\varepsilon/\partial T)_\rho$.

We could write down the dispersion relation for this coupled wave problem in terms of a three by three determinant in k^2 and ω^2. For many practical situations we again assume a quasi steady-state solution for θ_1^* and ρ_1^* proportional to $E_L^* E_s$ and substitute these values into the equation for E_s. If the terms in $(\partial\varepsilon/\partial T)_\rho$ are ignored, the resulting gain constant for the intensity

may be expressed in the form

$$g_s = -\frac{\omega_s}{n_s c}|A_L|^2$$

$$\times \operatorname{Im}\left[\frac{(\partial\varepsilon/\partial\rho)T\alpha\beta n_s c(v_{ac}{}^2k^2/\gamma C_v)+\rho_0(\partial\varepsilon/\partial\rho)_T{}^2\,k^2(-\kappa k^2-i\omega)}{[-\omega^2+(v^2{}_{ac}k^2/\gamma)+2i\omega\tau_{ac}{}^{-1}]\,(-\kappa k^2-i\omega)+i(1-\gamma^{-1})\,\omega v_{ac}{}^2k^2}\right], \quad (48)$$

where $\omega = \omega_L - \omega_s$ and $k = |\mathbf{k}_L - \mathbf{k}_s|$. For $\alpha = 0$ and $\gamma = 1$, which holds for nonabsorbing, dense fluids, the expression reduces to the Brillouin gain given by eqn (46).

The physical significance can best be appreciated by considering separately the regions where the frequency shift of the scattered light is very small, and and where it is nearly equal to the Brillouin shift $\omega_B = v_{ac}\,k$.

(a) *Thermal Rayleigh Scattering*, $\omega \approx \Gamma_R = \kappa k^2$

In this case the denominator is resonant near zero frequency, and the gain constant is given approximately by,

$$g_R = -\frac{\omega_s}{n_s c}|A_L|^2\frac{\alpha\beta n_s c}{C_v}\left(\frac{\partial\varepsilon}{\partial\rho}\right)_T\frac{\omega}{\omega^2+\Gamma_R{}^2}. \quad (49)$$

Note that Γ_R measures the width of the Rayleigh line, and that the gain constant is positive for negative ω. This means that the stimulated thermal Rayleigh scattering exhibits an antistokes shift. Light of a higher frequency than that of the incident laser beam is generated. One would get the normal stokes shift, if the electrocaloric effect alone were operative. In the absence of absorption, only the terms $(\partial\varepsilon/\partial T)_\rho$ which were ignored in eqn (48) would be active. The normal stokes shift also occurs in the Rayleigh wing scattering from anisotropic molecules. This is to be expected on thermodynamic grounds. In the case of absorption there is another source of energy available. The thermal energy makes conversion to higher energy photons possible. Experimentally the width is not determined by Γ_R which is usually very small, but rather by the line width of the laser source or by transient phenomena.

(*b*) *Thermal Brillouin Scattering*, $\omega \simeq \omega_B = v_{ac}k$.

In this case the last term in the numerator of eqn (48) leads to the normal Brillouin gain, given by eqn (46). To this must be added a correction from the thermal effect

$$g_{TB} = \frac{\omega_s|A_L|^2}{n_s c}\left(\frac{\partial\varepsilon}{\partial\rho}\right)_T\frac{\alpha\beta n_s c}{C_v}\frac{\tau_{ac}{}^2\omega_B{}^2}{2\omega^2}\frac{(\omega-\omega_B)}{1+\tau_{ac}{}^2(\omega-\omega_B)^2}. \quad (50)$$

This expression is positive for $\omega_L - \omega_s - \omega_B = \omega - \omega_B > 0$. There is more gain above the exact downward Brillouin frequency shift ω_B. This "dispersive" behaviour of the gain is shown in Fig. 3c. There is very good agreement between theory [17] and experiment.[16]

This absorptive effect could be superimposed equally well on the stimulated Raman effect. The thermal expansion coefficient β must then be replaced by a constant which describes the thermal variation of the optical phonon frequency, or of $(\partial\alpha/\partial Q)$.

The derivation given here applies only to a quasi steady-state. Transient effects during very short laser pulses will show a considerably different behaviour, since thermal relaxation and thermal expansion processes may not have time to develop.

10. Stimulated Raman Scattering from Polaritons

In ionic piezoelectric crystals the optical phonon modes are directly coupled to an electromagnetic mode with the same frequency and wave number. A mixed mode results, which is simultaneously infrared and Raman active. This coupling of the ionic and electronic oscillators results in a polariton wave which has both electromagnetic and vibrational character. When a laser beam is incident on such a crystal it is possible to generate the stokes frequency equal to the laser frequency minus the polariton frequency. The coupling coefficient is an interference of a Raman-type coupling and a parametric coupling between three electromagnetic waves. As Shen[18] first pointed out, the simultaneous coupling of the wave at the stokes frequency $\omega_2 = \omega_L - \omega_1$, the optical phonon at ω_1 which has an uncoupled eigenfrequency ω_0, and the infrared electromagnetic wave at ω_1 must be considered,

$$\nabla^2 E_2 + \varepsilon_2(\omega_2/c)^2 E_2 = -4\pi(\omega_2/c)^2(N d_Q E_L Q^* + N d_E E_L E_1^*), \quad (51a)$$

$$\nabla^2 E_1^* + \varepsilon_\infty(\omega_1/c)^2 E_1^* = -4\pi(\omega_1/c)^2(N d_E^* E_L^* E_2 + N e Q^*), \quad (51b)$$

$$-\mu\omega_1^2 Q^* + i\omega\Gamma\mu Q^* + \mu\omega_0^2 Q^* = e E_1^* + d_Q^* E_L^* E_2. \quad (51c)$$

Here μ is the reduced mass of the ionic oscillator with an effective charge e. The quantity d_Q is equivalent to the variation of the polarization with the ionic coordinate, which was called $\partial\alpha/\partial Q$ in eqns (27–32) of Section 6. We have ignored the dispersion term proportional to β in the optical vibration. The quantity d_E is equivalent to the nonlinear susceptibility used in eqns (19).

Assume exponential solutions for the three fields of the form

$$E_2 = A_2 \exp(i\mathbf{k}_2 \cdot \mathbf{r} + \gamma_2 \cdot \mathbf{r})$$

$$E_1 = A_1 \exp(i\mathbf{k}_1 \cdot \mathbf{r} + \gamma_1 \cdot \mathbf{r})$$

$$Q = Q_1 \exp(i\mathbf{k}_1 \cdot \mathbf{r} + \gamma_1 \cdot \mathbf{r})$$

with $\mathbf{k}_1 = \mathbf{k}_L - \mathbf{k}_2$ and $\gamma_1 = \gamma_2$. Substitution into the three coupled equations leads to a secular equation, from which γ can be determined. (There can only be gain in the direction normal to the crystal boundary.)

$$\begin{vmatrix} 4\pi d_E A_L & (d_Q/e)A_L & \varepsilon_2 - (\omega_2/c)^2(\mathbf{k}_2 - i\gamma)^2 \\ \varepsilon_\infty - (c^2/\omega_1{}^2)(\mathbf{k}_1 + i\gamma)^2 & 1 & 4\pi d_E A_L{}^* \\ -1 & \mu(-\omega_1{}^2 + \omega_0{}^2 + i\omega\gamma)/4\pi N e^2 & -(d_Q/e)A_L{}^* \end{vmatrix} = 0 \tag{52}$$

For the detailed algebraic solutions we refer to the literature.[18] The following approximation is, however, very good in the neighbourhood of the polariton resonance. Here the vibration damping Γ is dominant. One solves the second equation for $E_1{}^*$, substitutes into the equation for Q^* and solves for Q^* in terms of $E_L{}^* E_2$. Substitution into the first equation then leads to an exponential gain for the wave E_2. The result is

$$2(k_2 - \varepsilon_2{}^{1/2}\,\omega_2\,c^{-1}) - 2i\gamma = \frac{4\pi N e \omega_2{}^2}{\mu k_2\,c^2}\,|A_L|^2 \left(\frac{\mu(\omega_0{}^2 - \omega_1{}^2)}{Ne}\,d_E + d_Q\right)^2$$

$$\times \left[\left(\omega_0{}^2 - \frac{4\pi N e^2/\mu}{(k_1{}^2 c^2/\omega_1{}^2) - \varepsilon_\infty} - \omega_1{}^2\right) - i\omega_1\Gamma\right]^{-1}. \tag{53}$$

The last term contains a resonant denominator. Its real part vanishes for

$$k_1{}^2 = \frac{\omega_1{}^2}{c^2}\left[\varepsilon_\infty + \frac{4\pi N e^2}{\mu(\omega_0{}^2 - \omega_1{}^2)}\right]. \tag{54}$$

This is precisely the dispersion curve for the undamped polariton. If there is momentum matching between the laser wave, the stokes wave and the undamped polariton, the gain is maximum. The gain constant is proportional to

$$\left(d_E + \frac{N e d_Q}{\mu(\omega_0{}^2 - \omega_1{}^2)}\right)^2,$$

which describes the interference of the direct electronic parametric coupling and the Raman type coupling via the ionic vibrations, which is resonant for $\omega_1 = \omega_0$. The gain constant itself does not exhibit resonant behaviour at this frequency. As the ionic vibration is excited, there is a concomitant electromagnetic radiation component. Because of the strong absorption, the power flux density at this infrared frequency is small compared to the flux density at the stokes frequency. The ratio is about $g_2\omega_1/\omega_2\alpha$, where g_2 is the power gain constant at ω_2 and α the absorption coefficient at ω_1.

The variation of the coupling parameter when the difference frequency of the two visible light waves goes through the ionic infrared resonance has been

observed in GaP, when visible and infrared laser beams were mixed. The stimulated Raman scattering from polaritons has been observed[19] in $LiNbO_3$. The stokes shift of the Raman scattering in the forward direction is 497 cm^{-1}, while in the backward direction it is 631 cm^{-1}. In this last case the momentum of the wave corresponds to an essentially pure TO phonon. In the former case the dispersion curve is cut at an appreciably lower frequency of mixed vibrational character. The dispersive behaviour of two A_1 symmetry modes in $LiNbO_3$ is shown in Fig. 4.

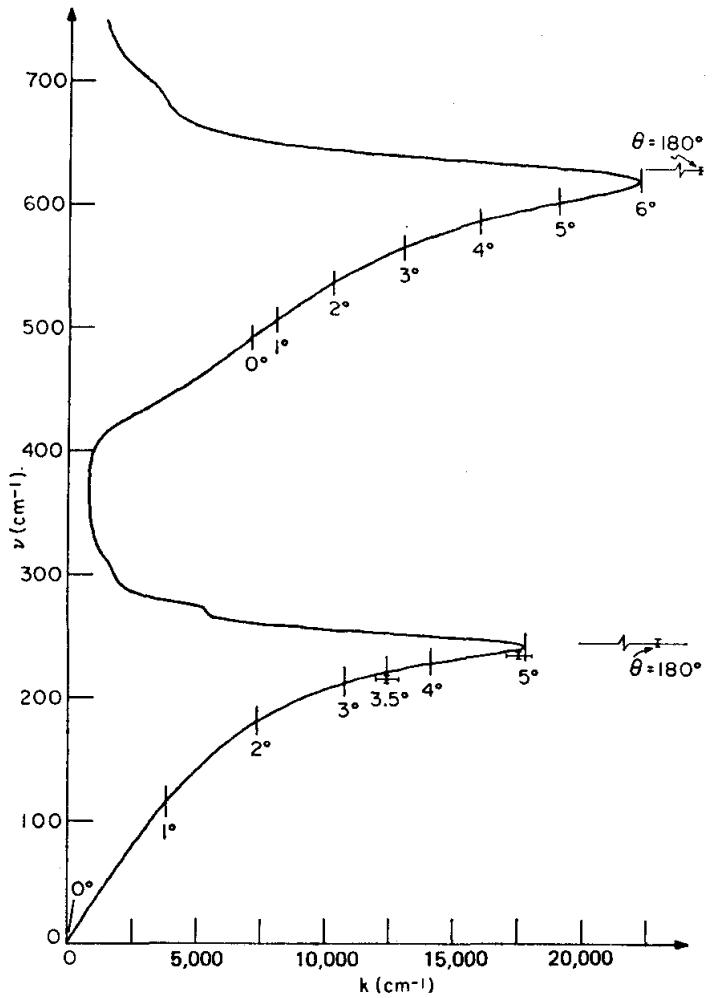


Fig. 4. The polariton dispersion curves characteristic of A_1 symmetry modes in $LiNbO_3$. The scattering angle θ is the angle between the incident laser light and the stokes radiation. The conditions for energy and momentum matching are indicated for Raman scattering of ruby laser light. (After Gelbwachs *et al.*[19]).

The complications which arise when more optical phonon branches are present, or when account should be taken of the different modes of polarization, will not be reviewed here.

11. Coupling of Light Waves with other Excitations

The pattern that has been established in preceding sections can be extended to the coupling between all types of classical waves, which correspond to boson excitations. These include plasmons, spin waves, helicons, rotons etc. All these waves are, of course, not only coupled to the light waves, but are also coupled to each other. The spin waves are, for example, strongly coupled to acoustic waves through the magnetoelastic coupling. Plasmons in semiconductors can also be strongly coupled with infrared phonons. If the coupled wave problem is linearized, the complex propagation constants with corresponding gain (or loss) may be obtained from a secular determinantal equation. The physical nature of the coupling is, of course, different for each pair of waves.

Here only the coupling of a light wave with a plasma wave and with a spin wave will be written down as further examples.

a. Plasma Raman scattering

Consider a small volume element at the point $\mathbf{r}$. Let the average deviation of the electrons from their equilibrium position in this volume element be $\mathbf{u}(\mathbf{r})$. Introduce normal coordinates $\mathbf{Q}_\mathbf{k}$ as the Fourier transform of this average deviation or local strain of the electron gas,

$$\mathbf{Q}_\mathbf{k} = \int \mathbf{u}(\mathbf{r}) \exp(-i\mathbf{k} \cdot \mathbf{r}) \mathrm{d}^3 r.$$

The canonical conjugate to this variable is $\mathbf{P}_\mathbf{k}$. The Hamiltonian density for the plasma waves then takes the form,

$$\mathscr{H}_{\text{plasma}} = \tfrac{1}{2} \sum_k [(1/Nm)\mathbf{P}_\mathbf{k} \cdot \mathbf{P}_{-\mathbf{k}} + \alpha k^2 \mathbf{Q}_\mathbf{k} \cdot \mathbf{Q}_{-\mathbf{k}} + 4\pi N^2 e^2 \mathbf{Q}_\mathbf{k} \cdot \mathbf{Q}_{-\mathbf{k}}]. \tag{55}$$

Here N is the average number of electrons per unit volume and α is the bulk modulus of the electron gas. The fluctuation in the electron density from the average due to the presence of plasma waves is

$$\delta\rho(\mathbf{r}) = N \operatorname{div}\mathbf{u} = iN \sum_k \mathbf{k} \cdot \mathbf{Q}_\mathbf{k} \exp(i\mathbf{k} \cdot \mathbf{r}).$$

The change in the interaction of the two light waves with the electrons in a unit volume due to the presence of the plasma waves is consequently

$$\mathscr{H}_{\text{int}} = (e^2/2mc^2)\mathbf{A}^2 \delta\rho(\mathbf{r}),$$

where

$$\mathbf{A} = \mathbf{A}_L \exp(i\mathbf{q}_L \cdot \mathbf{r} - i\omega_L t) + \mathbf{A}_s \exp(i\mathbf{q}_s \cdot \mathbf{r} - i\omega_s t) + \text{c.c.}$$

When all nonresonant perturbations are truncated, the interaction Hamiltonian density between the two linear parallel polarized light waves and the longi-

tudinal plasma waves ($\mathbf{Q}||\mathbf{k}$) becomes,

$$\mathscr{H}_{\text{int}} = (iNe^2/mc^2)\sum_k kA_L\, A_s^* Q_k^* \exp\left[i(\mathbf{q}_L - \mathbf{q}_s - \mathbf{k})\,.\,\mathbf{r}\right] + \text{c.c.} \qquad (56)$$

The equations of motion for the plasma coordinate are

$$\dot{P}_k = -\partial(\mathscr{H}_{\text{plasma}} + \mathscr{H}_{\text{int}})/\partial Q_k,$$

$$\dot{Q}_k = +\partial(\mathscr{H}_{\text{plasma}} + \mathscr{H}_{\text{int}})/\partial P_k.$$

These equations of motion can be combined into a wave equation for Q_k. Because of the presence of $\mathscr{H}_{\text{int}}$, a driving term proportional to the light amplitudes $A_L A_s^*$ is added to the plasma wave equation. Landau damping and damping by collisions may be taken into account by a phenomenological damping term. The wave equation for Q_k is

$$\ddot{Q}_k + \alpha\nabla^2 Q_k + \omega_p{}^2 Q_k = (ie^2k/m^2c^2)A_L\, A_s^* + (2i\omega_k/\tau')\, Q_k. \qquad (57a)$$

The wave equations for the light amplitudes A_L and A_s are also augmented by a nonlinear term, because the interaction Hamiltonian gives rise to a nonlinear current density,

$$j^{NL}(\omega_s) = -c\partial\mathscr{H}_{\text{int}}/\partial A_s^* = (+iNe^2/mc)\, kA_L\, Q_k^*$$

and a similar expression for $j^{NL}(\omega_L)$. The wave equations for the two light waves become, consequently,

$$-\ddot{A}_L + c^2\nabla^2 A_L = (4\pi iNe^2/m)\, kA_s\, Q_k, \qquad (57b)$$

$$-\ddot{A}_s + c^2\nabla^2 A_s = (4\pi iNe^2/m)\, kA_L\, Q_k^*. \qquad (57c)$$

The set of three coupled nonlinear wave equations is familiar from the Brillouin and Raman effect in other media.

b. Spin wave Raman scattering.[20]

The dominant physical mechanism for the coupling is via the spin-orbit interaction. Suppose the ground state of an ion is an orbital s-state, with $m_L = 0$, and a spin $m_s = \pm\frac{1}{2}$. Consider an excited p-state with a spin-orbit coupling. It admixes the states with $m_L = 0$, $m_s = \frac{1}{2}$ with $m_L = 1$, $m_s = -\frac{1}{2}$. A light wave with a circular polarization around the direction of quantization $\hat{z}$ of the spin wave will have an electric dipole matrix element

$$\langle s, m_L = 0,\ m_s = -\tfrac{1}{2}|er^+ E_1|p,\ m_L = 1,\ m_s = -\tfrac{1}{2}\rangle.$$

Another light wave with a linear component in the $\hat{z}$ direction will have a dipole matrix element

$$\langle p, m_L = 0,\ m_s = \tfrac{1}{2}|ezE_2|s,\ m_L = 0,\ m_s = \tfrac{1}{2}\rangle.$$

Since the two intermediate states are linked by the spin-orbit coupling, it is thus possible for the two light waves to produce a spin flip in the ground state. Very crudely, the value of the matrix element for the spin Raman effect will be lowered by the ratio of the spin-orbit coupling parameter to the orbital splitting. The two light waves will drive the spin excitation on each ion site with a phase determined by the wave vector difference $\mathbf{q} = \mathbf{k}_2 - \mathbf{k}_1$. They are thus driving a spin wave of wave vector $\mathbf{q}$ and frequency $\omega_2 - \omega_1$. Significant coupling will only occur, if both energy and momentum matching can be satisfied. The equation for the light driven spin wave is

$$\left[i\frac{\partial}{\partial t} - \omega_0 + i\Gamma + \beta\nabla^2 \right] \langle S_q(\mathbf{r}, t) \rangle = \lambda E_2 E_1^*. \tag{58}$$

In combination with the wave equation for the stokes wave E_1, the parametric gain of these waves pumped by the laser field E_2 may be derived in the usual manner.

These couplings have not yet been observed as stimulated effects. Spontaneous Raman scattering has, however, been studied from a wide variety of excitations. These include localized impurity vibrations, localized impurity spin modes, light scattering with the creation of two spin waves, two phonons, two rotons in liquid helium II, with a vibrational excitation in solid orthohydrogen, concentration fluctuations in fluid mixtures, etc. The quantum description of these spontaneous scattering processes is outside the scope of this review.

12. Transient Response

The discussion has so far been restricted to steady state type solutions. In most cases we have assumed monochromatic waves growing in space. Pure temporal growth can be treated in the same manner, as was shown by the fact that eqns (30) have the same form as eqns (25). A combined growth in space and time requires, however, the solution of coupled *partial* differential equations of the form given by eqns (19) and (20). Such solutions are of practical importance, because the powerful laser pump is often a short travelling pulse. The pulse duration may be quite short compared to the characteristic damping times. Consider for example the case of the stimulated Raman effect, when the excitation is by a mode-locked picosecond pulse laser train. The life time of the vibrational-rotational state of the molecule will often be longer than 10^{-12} sec. This is certainly so in a gas. One has a kind of shock excitation of each individual molecule, but this excitation does not propagate, as the velocity of the vibrational wave is small. The stokes light will however travel in synchronism with the laser pulse, as long as there is no dispersion in the group

velocities. We thus have a spatial growth of the stokes light, coupled with a pure temporal growth of the vibration. The gain in this situation must be derived, not from eqns (30), but from the coupled partial differential equations,

$$v_s \frac{\partial A_s}{\partial z} + \frac{\partial A_s}{\partial t} + \frac{A_s}{\tau_s} = c_1 A_v^*, \tag{59a}$$

$$v_v \frac{\partial A_v^*}{\partial z} + \frac{\partial A_v}{\partial t} + \frac{A_v}{\tau_v} = c_2 A_s. \tag{59b}$$

The constants c_1 and c_2 are shorthand notations for the expressions that occur on the right hand side of eqns (30). The laser amplitude is considered to be a constant parameter. The constants are related to the pure spatial Raman power gain constant g_s given by eqn (24) as follows; $2c_1c_2\tau_v = g_s v_s$. Rather general solutions of the set of eqns (59) can be found. They have the same form as the current and voltage equations on a transmission line. Transient pulses on such lines have been studied extensively. Solutions which are applicable to the stimulated Brillouin and Raman scattering have been given by Kroll[21] and by Wang.[22]

To minimize the algebraic complexity while retaining the essential features, let us assume that the damping τ_s^{-1} of the light wave and the group velocity of the vibrational wave are negligible. The substitution $x = z + v_s t$ then transforms the equations to

$$\frac{\partial A_s}{\partial x} = c_1 v_s^{-1} A_v^*,$$

$$\frac{\partial A_v^*}{\partial t} + \frac{A_v^*}{\tau_v} = c_2 A_s,$$

or

$$\frac{\partial^2 A_s}{\partial x\, \partial t} + \frac{1}{\tau_v}\frac{\partial A_s}{\partial x} - \frac{g_s}{2\tau_v} A_s = 0. \tag{60}$$

A_v^* satisfies the same equation. The damping term may be eliminated by the substitution $G = A_s \exp(t/\tau_v)$. A special solution of the hyperbolic equation

$$\frac{\partial^2 G}{\partial x\, \partial t} - \frac{g_s}{2\tau_v} G = 0$$

is the Bessel function of imaginary argument $J_0[i\sqrt{(2g_s x t/\tau_v)}]$. Solutions satisfying other initial and boundary conditions may be found by the Riemann procedure.[23]

Consider the case where the medium, continuously pumped by the constant laser field, has a very short stokes pulse as input. Before the pulse arrives at a certain volume element, there is no excitation of the molecular vibrational coordinate. The initial conditions in this case are

$$A_s(0, t) = G_0(t), \qquad (\partial A_s/\partial x)_{t=0} = 0. \tag{61}$$

Here $G_0(t)$ is the shape of the pulse at the entrance of the Raman cell.

The solution satisfying eqns (60) and (61) is

$$A_s(x, t) = G_0(t) - \int_0^t G_0(t') \exp\left[-(t-t')/\tau_v\right] \frac{\partial J_0\{i\sqrt{[2g_s(t-t')x/\tau_v]}\}}{\partial t'}\, dt' \tag{62}$$

For a short rectangular pulse of duration t_p, $G_0(t) = G_0$ for $0<t<t_p$ and vanishes outside this interval. The cell length satisfies the condition $l \gg v_s t_p$ and appreciable gain is assumed so that the argument $X = \sqrt{(2g_s t_p x/\tau_v)} \gg 1$. In this limit the following asymptotic expression may be used

$$\frac{\partial J_0(iX)}{\partial X} = -iJ_1(iX) \to e^X/(2\pi X)^{1/2}.$$

The major contribution to the integral (62) will come from values $t' \sim 0$, where the integrand is largest. In this approximation one finds

$$A_s = G_0(4\pi g_s x t/\tau_v)^{-1/2} \exp\left[(2g_s x t/\tau_v)^{1/2} - t/\tau_v\right]. \tag{63}$$

The leading edge of the pulse retains the value G_0 but the trailing edge, $t=t_p$, reaches its maximum value at the end of the cell $x=l$. It is seen that the transient gain is greatly reduced compared to the steady state value $g_s l$, and increases as the square root of the cell length. The solution given by eqn (63) is valid only for short times such that the gain dominates the loss in the exponent, i.e. for $t<2g_s l\tau_v$. The transient regime holds if the pulse duration is short compared to the life time of the vibrational state multiplied by the steady state gain. For times longer than this, the gain rapidly approaches its steady state value as follows from eqn (62) if an upper limit $t \to \infty$ is taken.

As the transient gain increases, the pulse becomes sharper in the time domain. It reaches half its maximum value at a time $t_p - \Delta t_{1/2}$, where

$$\exp\{2g_s l(t_p - \Delta t_{1/2})/\tau_v\}^{1/2} = \tfrac{1}{2} \exp\{2g_s l t_p/\tau_v\}^{1/2}$$

or

$$\Delta t_{1/2} \approx 2 \ln 2 (t_p/G_{\max}).$$

This sharpening in time implies, of course, a broadening of the stokes spectrum in frequency. This is just the opposite from what happens in the pure steady state Raman effect, where the line is narrowed since the maximum gain occurs at the centre of the resonance. A short input pulse implies a certain

frequency spread in the incident signal. As amplification takes place different frequency components in the vibrational and stokes waves will beat with each other. This leads to a gradual broadening, in a way which has some similarity to the self broadening caused by modulation of the intensity dependent index of refraction. The transient gain effect may thus lead to very short pulses. The limit is set by the dispersion of the group velocities if the laser pump is also a short pulse, but if the pump is a quasi-continuous long pulse, the fundamental limit appears to be a duration comparable to the inverse of the vibrational frequency itself.

For a short laser pump pulse, there is of course only gain for the stokes pulse travelling in synchronism in the forward direction. There is no significant gain for a backward travelling light pulse. The same considerations hold for the stimulated Brillouin effect, since the group velocity of the acoustic phonons is also negligible compared to the light velocities. Since the acoustic phonon damping times are often several orders of magnitude longer than the vibrational-rotational life times, the transient regime is first reached for the acoustic waves and the gain for Brillouin scattering is correspondingly reduced. Although the steady state gain for the Brillouin process may be larger than the steady state gain for the stimulated Raman process, the latter may dominate when the laser pulse duration satisfies the condition $G_{ac}\tau_{ac} > t_p > \tau_{vib}$. Such competition between nonlinear effects has also been observed.

The deformation in time of a laser pulse propagating in a medium with an intensity dependent index of refraction provides another example of a transient nonlinear effect. The self-focussing occurring in media with a quadratic Kerr effect must be extended to include the combined space–time behaviour. Here we shall briefly discuss the development in time of a spatially homogeneous plane wave pulse. Since the index of refraction is highest, when the intensity is highest, the maximum of the pulse will travel at a slower velocity than the rest. The pulse will thus get a steeper trailing edge, while the leading edge becomes more gradual.[24] Before the shape of the envelope is modified significantly in this fashion, the phase retardation, which is also a function of the intensity, will have an appreciable time dependence. This will lead to a characteristic observable frequency broadening of the spectrum of the pulse.[25,26] Our starting point is again the nonlinear complex amplitude equation

$$v_g \frac{\partial A}{\partial z} + \frac{\partial A}{\partial t} = \frac{i\omega_0}{2\varepsilon_0} \delta\varepsilon(t) A(z,t). \tag{64}$$

The nonlinear polarization corresponding to the quadratic Kerr effect in the steady state is given by

$$P^{NL}(\omega_0) = \chi^{NL}(-\omega_0, \omega_0, -\omega_0, \omega_0) |A|^2 = (\varepsilon_2/4\pi) |A|^2 A.$$

The temporal behaviour of $\delta\varepsilon(t)$ is governed by the Fokker–Planck equation for rotational relaxation discussed in Section 7. It approaches its instantaneous equilibrium value $\varepsilon_2|A|^2$ with a relaxation time τ,

$$\frac{\partial(\partial\varepsilon)}{\partial t} = -\frac{1}{\tau}(\partial\varepsilon - \varepsilon_2|A|^2). \tag{65}$$

If the pulse width $v_g t_p \ll z < l$ is very short compared to the cell length l, the time derivative on the left-hand side of eqn (64) may be neglected in comparison with the spatial derivative. Substitute the solution for $\delta\varepsilon(t)$ and separate eqn (64) into its real and imaginary parts writing the complex amplitude as $A = |A| \exp(-i\phi)$. The amplitude of the pulse $|A(t')|$ may be considered independent of z, if pulse steepening or pulse distortion due to dispersion of the group velocities is negligible. Under these conditions the phase ϕ is given by the simple expression

$$\phi(z, t) = \frac{k_0 z \varepsilon_2}{2\varepsilon_0} \int_{-\infty}^{t} \frac{1}{\tau} \exp[-(t-t')/\tau] \, |A(t')|^2 \, dt'. \tag{66}$$

The frequency components of the pulse having propagated a distance z are given by the Fourier transform

$$A(\omega, z) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} |A(t)| \exp[i(\omega - \omega_0) t] \exp[-i\phi(z, t)] \, dt. \tag{67}$$

The methods of stationary phase applied to this expression show that the frequency shift is essentially given by the slope of the phase curve $-\partial\phi/\partial t$. There are usually two points t_1 and t_2 on the phase curve with the same slope. These points can interfere constructively or destructively. If they have about equal amplitudes $|A(t_1)|$ and $|A(t_2)|$, the Fourier component will be maximum or near zero at some frequencies. If the relaxation time is very short, $-\partial\phi/\partial t$ is proportional to $d|A(t')|^2/dt'$ and the broadening would be symmetrical for an initially symmetrical pulse. For long relaxation times $-\partial\phi/\partial t$ is essentially proportional to $-|A|^2$ and the broadening should occur mostly on the low frequency side. All these features are consistent with detailed numerical calculations which evaluate $|A(\omega, z)|^2$ from the expressions (66) and (67) and are shown in Fig. 5. This typical spectral shape also agrees qualitatively with many experimental observations of the broadening of short pulses. The results are also consistent with the picture of repeated beating of Fourier components in the frequency domain. The effect can be described as a transient modulation of the index of refraction.

Finally, transient effects in thermal Rayleigh scattering should be mentioned. Some characteristic solutions of eqns (47) in Section 9 displaying

combined spatial–temporal behaviour have recently been compared with experimental results.[27] A remarkable feature is that there is a transient gain for zero frequency shift and that the gain initially increases quadratically with time.

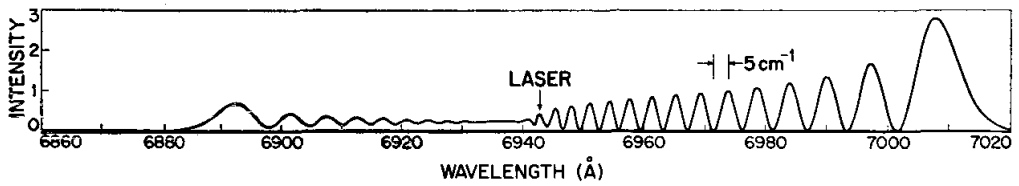


Fig. 5. The initial pulse has a Gaussian shape with a full width at $1/e$ level given by $\tau_p = 5\times10^{-12}$sec. The circular frequency ω_0 corresponds to the ruby laser wavelength. The relaxation time of the dielectric constant in CS_2 is taken as $\tau = 2\times10^{-12}$sec. The frequency broadening is calculated at a cell length corresponding to the position before appreciable envelope distortion has occurred. The intensity in arbitrary units of the spectrum is calculated from eqns (66) and (67), (After Gustafson *et al.*[26]).

Another interesting effect is forward thermal Rayleigh scattering induced by picosecond pulses.[28] It is clear that on this time scale density fluctuations have no time to develop. The effect can be understood in terms of eqns (47) as follows. The spatial and/or temporal beat of two incident laser pulses creates a temperature variation $\theta_1(\omega_L-\omega_L', k_L-k_L')$. This may be regarded as a steady state process, if the distribution of the absorbed power by the dye molecules to the surrounding solvent molecules is instantaneous. The density variations on this time scale of a few picoseconds are, however, negligible and one may take $\rho_1 = 0$, but a light wave at the new combination frequency $2\omega_L-\omega_L'$, and in the new direction $2k_L-k_L'$ can be driven by the term proportional to $(\partial\varepsilon/\partial T)_\rho\theta_1 E_L$ in eqn (47a). Many spatial and frequency side bands can thus be coupled together by the thermal Rayleigh scattering on this time scale. Even if the absorbed power is not immediately distributed over all degrees of freedom in the fluid, the effect would still be present. One should then take account specifically of the variation of the dielectric constant with the time varying occupation of various excited vibrational and rotational degrees of freedom. A number of characteristic constants and relaxation times would be needed to describe this in detail instead of the single constant $(\partial\varepsilon/\partial T)\rho$. The experimental results[28] confirmed that the coupling was proportional to the absorption coefficient of the dye solution, and to $(\partial\varepsilon/\partial T)_\rho$ characteristic of the solvent. Furthermore spatial side bands could be created on a probe beam at a frequency which was not absorbed by the dye. This is consistent with the picture of an index modulation proportional to $(\partial\varepsilon/\partial T)_\rho\theta_1$.

This cursory review is, of course, far from exhaustive. Its purpose has been to show how the extremely rich variety of nonlinear optical phenomena in condensed matter can be understood fairly well on the basis of a rather simple and unified classical point of view.

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Optical Pumping and Related Topics

G. W. SERIES

I. A PRELIMINARY SURVEY

1. INTRODUCTION

We propose to give an account of work which is commonly included under the titles:

(i) optical–radio frequency double resonance experiments,

(ii) level-crossing experiments,

(iii) optical pumping experiments.

For the most part, such experiments have been performed on free atoms, and the results that have been obtained are of interest in atomic spectroscopy in particular, and in atomic physics more widely, but the methods have also been applied to molecules and to ions in solids.

The experiments have in common that all the techniques are based on the interaction of light with the atoms, either in absorption or in emission or both. There are close connections with experiments in which electrons are used for excitation.

The light fields we encounter are weak: we shall not be concerned with non-linear effects, nor shall we be concerned with the statistical properties of the fields.

Historically, the experiment which introduced this whole field of work was the optical radio-frequency double resonance experiment of Brossel and Bitter.[1] This showed how it was possible to apply to excited atoms the techniques of magnetic resonance which had already been worked out for atoms in their ground states (atomic beam resonance methods).

We shall find it convenient to take the Brossel–Bitter experiment as our starting point, then to discuss level-crossing experiments, and then optical pumping. This is not the historical development. In the first section we shall aim to introduce the whole field, by discussing at length some simple examples. In later sections we shall take up subtle points and elaborate on techniques used in other experiments of the same types.

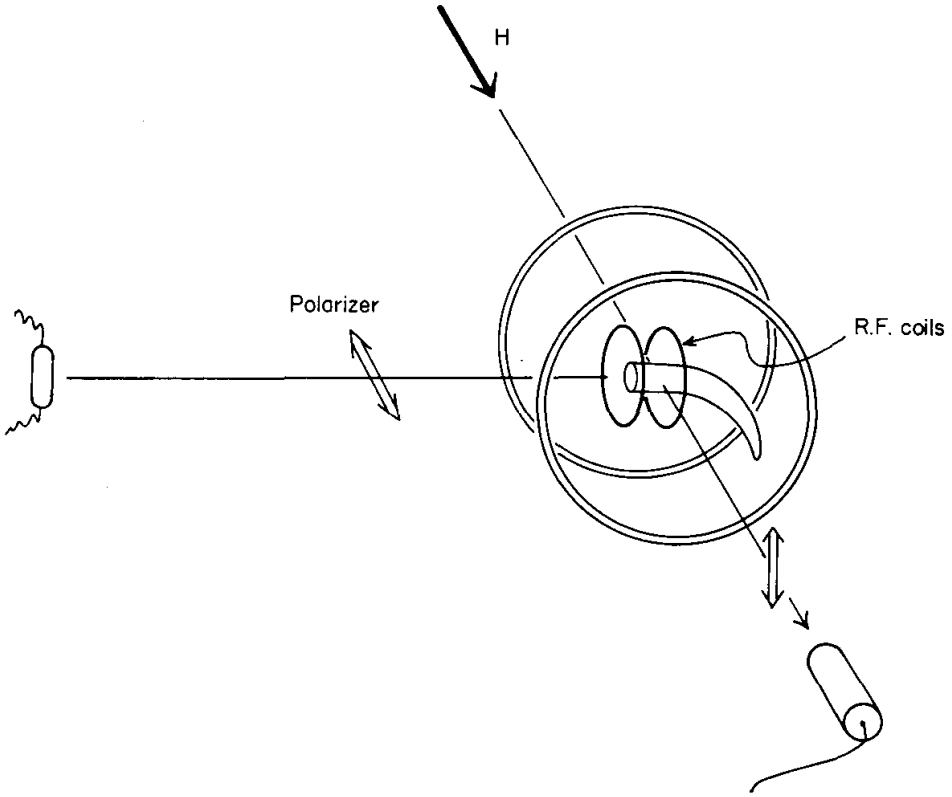


FIG. 1a. Schematic apparatus for performing the Brossel–Bitter experiment.

2. THE BROSSEL–BITTER EXPERIMENT

The Brossel–Bitter experiment measured the Zeeman splitting of the excited state $6s6p\ ^3P_1$ in mercury by magnetic resonance. The atoms are present in the form of a vapour at very low pressure in the horn-shaped vessel and irradiated by light from a mercury lamp (Fig. 1a). They undergo the process known as resonance fluorescence which can be described (crudely, but well enough for our immediate purpose) as the absorption of a photon, followed by the emission of a photon. The atoms remain in the excited state for a time determined by the probability of spontaneous emission (Einstein coefficient A). The mean lifetime is $\tau = A^{-1}$. This is the time available for the magnetic resonance transition.

In order to secure an unequal distribution of atoms over excited states m (Fig. 1b) the incident light is polarized. Magnetic resonance is stimulated by an oscillating magnetic field $H_1 \cos \omega_0 t$ produced by a current in the coils shown in Fig. 1a. The oscillating field is at right angles to the static field H which produces the splitting of levels shown in Fig. 1b. This geometrical arrangement stimulates transitions $\Delta m = \pm 1$.

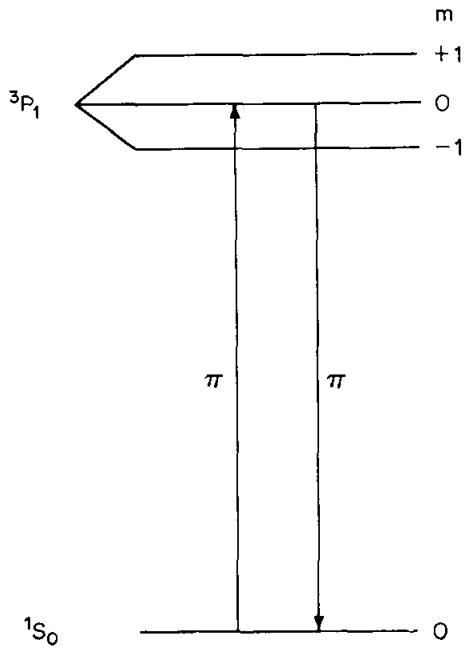


FIG. 1b. Resonance fluorescence in mercury stimulated by π-polarized light.

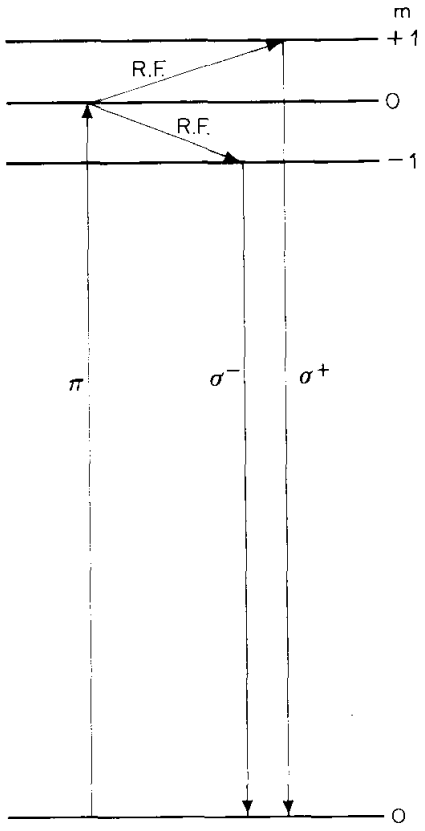


FIG. 1c. Change of polarization of fluorescent light resulting from radio-frequency transitions.

Atoms undergo these transitions:

(a) if the amplitude H_1 is sufficiently strong, and

(b) if the frequency ω_0 is sufficiently close to the splitting in the field H.

The resonance is detected by making use of the profound effect which the transitions have on the polarization of the emitted light: compare Figs. 1b and 1c, and notice how, as a result of a transition, the polarization is changed from π to σ. These have different spatial distributions. A photomultiplier in a fixed position registers the change. Thus the absorption or emission of a quantum of energy at the radio-frequency determines the polarization of an emitted quantum of vastly greater energy at the optical frequency. That is why this optical method of detection is so much more efficient, when one comes to consider questions of signal-to-noise ratio, than a direct measurement of the absorption of radio-frequency energy.

Figure 2 is an oscilloscope display of a Brossel–Bitter resonance. It was

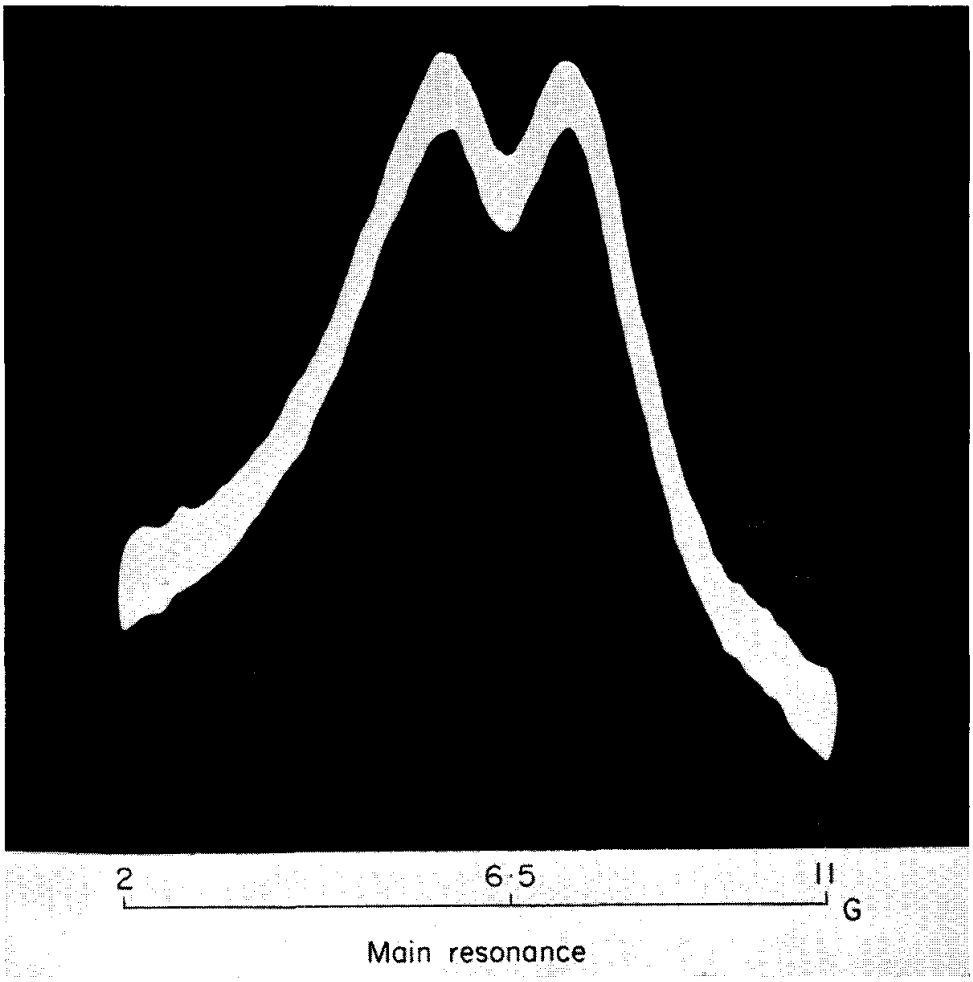


FIG. 2. Double resonance signal in the 6^3P_1 state of mercury.

obtained by keeping the frequency fixed at about 6 MHz and sweeping the magnetic field through the resonance region. It is worth noticing that this curve was obtained with very simple methods of detection: wideband amplification of the voltage developed across a resistor in the output of the photomultiplier.

The curve is double-peaked because the transitions $\Delta m = \pm 1$ are closely coupled. It is similar to the resonance curve of two over-coupled resonant circuits. The curve becomes single peaked when the radio-frequency field strength is reduced.

We shall shortly show how the shape of such resonance curves can be calculated. First, we make some remarks on the properties of the oscillating field required to stimulate the resonances. Our considerations here are not limited to the Brossel–Bitter experiment in particular; we have in mind this type of experiment in general.

2.1 AMPLITUDE OF OSCILLATING FIELD

It is important to consider what amplitude of radio-frequency field is necessary to induce transitions. A classical interpretation of magnetic resonance suggests that the field should be strong enough for the excited atom to precess through an angle of about one radian within its lifetime, τ. Recalling that the precessional velocity in the field H_1 is γH_1, where γ is the gyromagnetic ratio, we have for the required condition

$$(\gamma H_1)\,\tau \approx 1 \text{ rad.} \tag{1}$$

More formally, we find the factor $\langle J, M|J_x \pm iJ_y|J, M \mp 1\rangle$ on the left-hand side. Using typical values for τ (10^{-8}sec) and a value of γ corresponding to the Bohr magneton (1·4 MHz/G) we arrive at $H_1 \approx 10$ G. Actually, this is an over-estimate because we can detect changes of angle smaller than 1 rad, but it is clear we must think in terms of amplitudes of the order of a few gauss. Moreover, in order that the Zeeman levels be non-degenerate, we must think in terms of frequencies of the order of 10^8 rad sec^{-1}, i.e., in the range 10–100 MHz.

Clearly, in order to drive such a resonance we need high frequency oscillators with power amplifiers. These requirements stem immediately from the fact that we are dealing with excited states. At a later stage, when we come to consider resonances in the ground states, we shall find that the longer lifetimes allow the use of fields of the order of milligauss, and that kilocycle frequencies are often appropriate.

The use of high power, high frequency fields in studying resonances in excited states often raises technical problems which we shall discuss later.

2.2. WIDTH OF RESONANCE CURVES

An important characteristic of a resonance curve is its width. In the present case we can derive a minimum figure for the width by thinking in terms of Fourier transforms. The time available for the oscillating field to interact with the atom is limited by the mean lifetime of the excited state, hence the width of the resonance must be of order τ^{-1}. In quantum terms the point to notice is that the excited energy levels of Fig. 1b are broadened by radiation damping, so that the width of a resonance between a pair of levels cannot be less than the sum of their widths, $2\Gamma = 2\tau^{-1}$.

This is a minimum width. It may be increased by radio-frequency power broadening (shortening of the lifetime through transitions induced by the oscillating field) or by inhomogeneous static fields. The latter is not a serious problem for excited state resonances, though it is for ground states.

Of course, a precise definition of the width of a resonance curve rests upon knowledge of the profile.

2.3. ANALYTICAL EXPRESSION FOR THE LINE PROFILE

The following is a version of the calculation given by Brossel and Bitter. In the interests of simplicity, let us make the system effectively two-level by supposing that we are detecting the light through a circular polarizer (σ^+) in front of the photomultiplier. Then we are concerned only with transitions 0 to $+1$.

Suppose an atom is excited at time t_0. (This is the "pulse" approximation. It is valid if the spectral distribution of the exciting light is much greater than the natural width of the excited states. We shall frequently use this approximation, and discuss it in a later section).

Let $P(t, t_0)$ be the probability that, at time t, the atom has made a transition. This can be calculated from the theory of magnetic resonance. The result is

$$P(t, t_0) = (b^2/p^2) \sin^2 \tfrac{1}{2} p(t-t_0), \tag{2}$$

where

$$b = \gamma H_1; \qquad p = \gamma[(H-H_0)^2+H_1{}^2]^{\frac{1}{2}}; \qquad H_0 = \omega_0/\gamma;$$

H is the static field and $H_{\text{r.f.}} = 2H_1 \cos \omega_0 t$ the oscillating field. (Only one rotating component of $H_{\text{r.f.}}$ is effective: we are ignoring the counter-rotating component).

The probability that the atom has survived in the interval $(t-t_0)$ is exp $[-\Gamma (t-t_0)]$, where Γ is the decay constant for the excited states (the same for both). To find the probability of emission of a photon of different polarization, eqn (2) must be multiplied by this exponential factor.

If atoms are excited at the uniform rate R, the number excited in time $\mathrm{d}t_0$ is $R\,\mathrm{d}t_0$. Hence, finally, if the atoms are excited independently of one another,

the steady-state intensity of light reaching the detector is

$$I(H) \propto \int_0^t (b^2/p^2) \sin^2 \tfrac{1}{2}p(t-t_0) \exp[-\Gamma(t-t_0)]\, dt_0$$

$$\propto b^2/(p^2+\Gamma^2)$$

$$= (\gamma H_1)^2/\{\Gamma^2+[\gamma(H-H_0)]^2+(\gamma H_1)^2\} \tag{3}$$

This is a Lorentzian curve of full width at half-intensity, $2[H_1{}^2+(\Gamma/\gamma)^2]^{\frac{1}{2}}$. As H_1 tends to 0 the width tends to 2Γ (in radians $\sec^{-1}$). The power-broadening is thus quantitatively expressed.

In Brossel and Bitter's paper the probability $P(t, t_0)$ of equation (2) is replaced by the more complicated expression representing a transition out of the state 0, leading to a more complicated version of (3).

2.4. DOUBLE RESONANCE LINE WIDTH COMPARED WITH OPTICAL LINE WIDTH

The quantitative remarks we made earlier about line widths are borne out by these calculations and exemplified by Fig. 2. The line width of a few gauss in that figure corresponds to a frequency of the order of 1 MHz and a lifetime of order 10^{-7}sec. This line width should be compared with the line width in optical studies of the Zeeman effect—typically, several hundred gauss. The Brossel–Bitter experiment therefore allows the measurement of Zeeman intervals in excited states with a precision orders of magnitude better than optical measurements.

The method is not limited to the measurement of Zeeman intervals. We shall indicate in the next section how it has been applied to determine hyperfine structures.

3. Double Resonance Method: Hyperfine Structures

The basis of the double resonance method is to make use of the changes of polarization of fluorescent light which result from magnetic resonance transitions $\Delta M = \pm 1$. This selection rule stems from the existence of matrix elements of the $(x-y)$ component of the magnetic moment operator

$$\hat{\boldsymbol{\mu}} = (\beta/\hbar)(\hat{\mathbf{L}}+2\hat{\mathbf{S}}).$$

In the Brossel–Bitter experiment the states were the eigenstates $|M\rangle$ of $\hat{J}_z = \hat{L}_z+2\hat{S}_z$ and $\hat{\mu}_{xy}$ has non-vanishing matrix elements between these states. The static magnetic field was used only to remove the degeneracyof the $|M\rangle$.

In an atom having hyperfine structure with weak field quantum numbers F, M_F, the operator $\hat{\mu}_{xy}$ has matrix elements between states $|F, M_F\rangle$ and

$|F', M_F \pm 1\rangle$, where $F' - F = 0, \pm 1$. The polarization of the light is determined by the value of M_F, irrespective of whether levels of different M_F are degenerate. Hence, the double resonance method may be applied to measure hyperfine intervals in zero field ($F' - F = \pm 1$). It is also, of course, still applicable in fields strong enough to decouple I and J, and in intermediate fields. We shall illustrate this by reference to an experiment of Ritter and Series[2] on an excited state of potassium.

3.1. HYPERFINE RESONANCES IN ZERO FIELD

The apparatus is shown in Fig. 3. It is, in essentials, the same as that used by Brossel and Bitter, but differs in points of experimental technique. Most significant is that the potassium is introduced into the resonance region in the form of a crude atomic beam. The use of a hollow cathode light source is also noteworthy.

The excited states of interest here were the second excited P states $5^2P_{3/2}$ whose hyperfine structure allows a determination of the nuclear electric quadrupole moment. The level scheme is shown in Fig. 4a. A blue filter eliminates

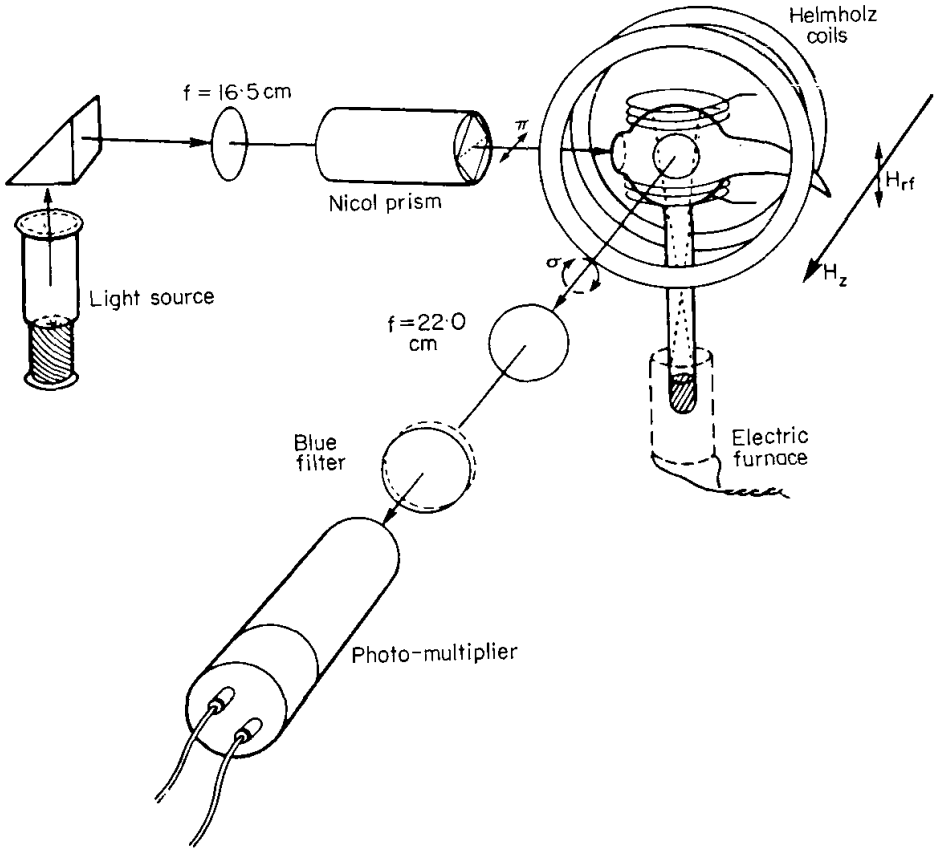


FIG. 3. Double resonance experiment to study the hyperfine structure of $5^2P_{3/2}$ of potassium. (After Ritter and Series.[2])

the much stronger fluorescence from the 4^2P states. Fluorescence from $5^2P_{1/2}$ passes the filter but its intensity is unchanged by the magnetic resonance unless circular polarizers are used (see later).

The hyperfine structure of $5^2P_{3/2}$ is shown in Fig. 4b. Magnetic resonance curves obtained by varying the frequency while keeping the amplitude H_1 fixed are shown in Fig. 5. The interpretation of these curves is not easy without

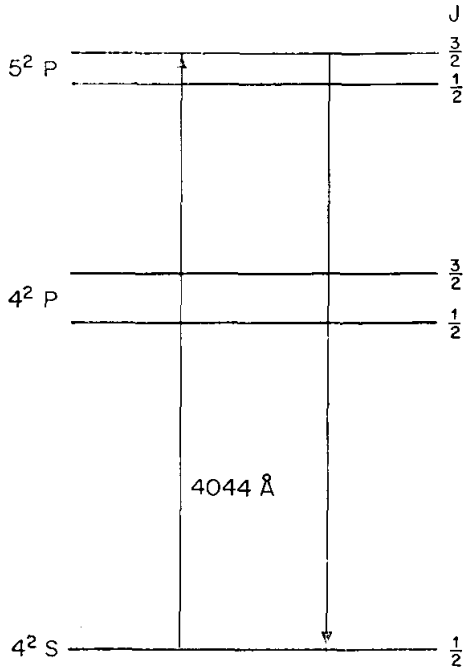


FIG. 4a. Resonance fluorescence of the second excited state in potassium.

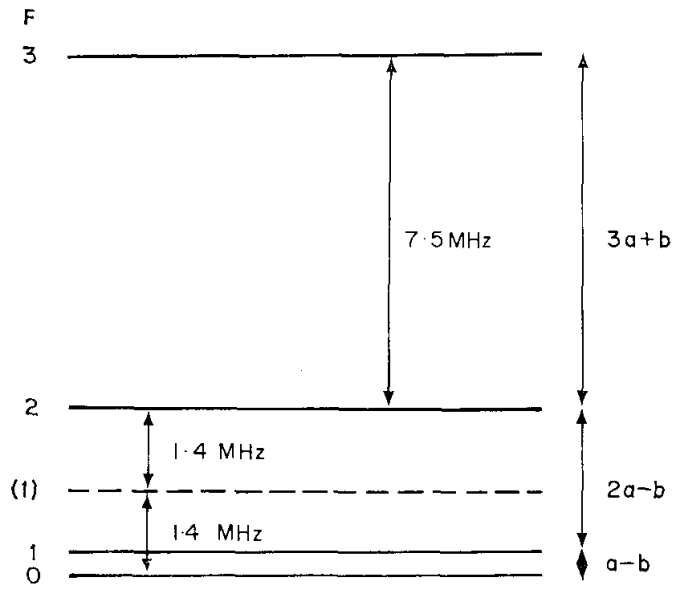


FIG. 4b. Hyperfine structure of $5^2P_{3/2}$, showing the double resonance transitions.

additional information (based partly on the known h.f.s. of the ground state and partly on analysis of double resonance curves in strong magnetic fields). With this information one can be confident that the curve at 7·5 MHz represents the complex of transitions between the various M_F levels belonging to $F = 3$ and $F = 2$. Notice that the width of the line at half intensity is about 2 MHz.

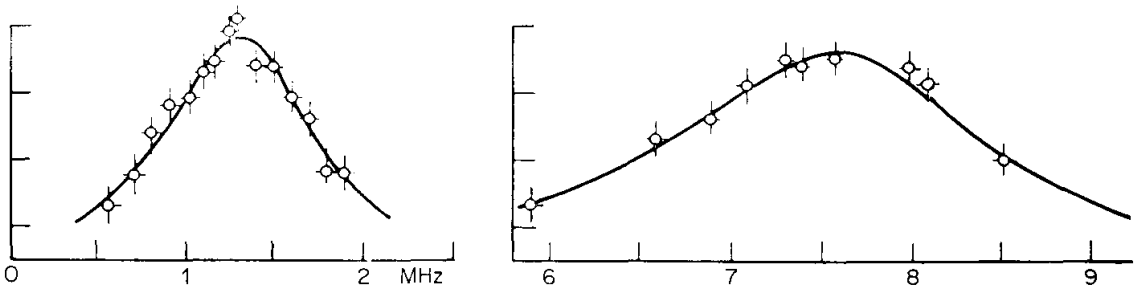


FIG. 5. Experimental curves corresponding to the transitions marked in Fig. 4b. (After Ritter and Series.[2])

The resonance curve at 1·4 MHz is obviously narrower and its interpretation is not immediately obvious. It is, in fact, identified by its width and position as a double-quantum transition between the levels $F = 0$ and 2. Such transitions are found when the separate resonances $0-1$ and $1-2$ overlap and the r.f. field is sufficiently strong. We shall discuss these multiple-quantum resonances later.

3.2. HYPERFINE STRUCTURE OF RESONANCES IN STRONG FIELD

" Strong field " is, of course, a relative term. The fact that 25 G is a strong field in this context (decouples I and J) emphasises the smallness of the hyperfine structures which the method is capable of measuring.

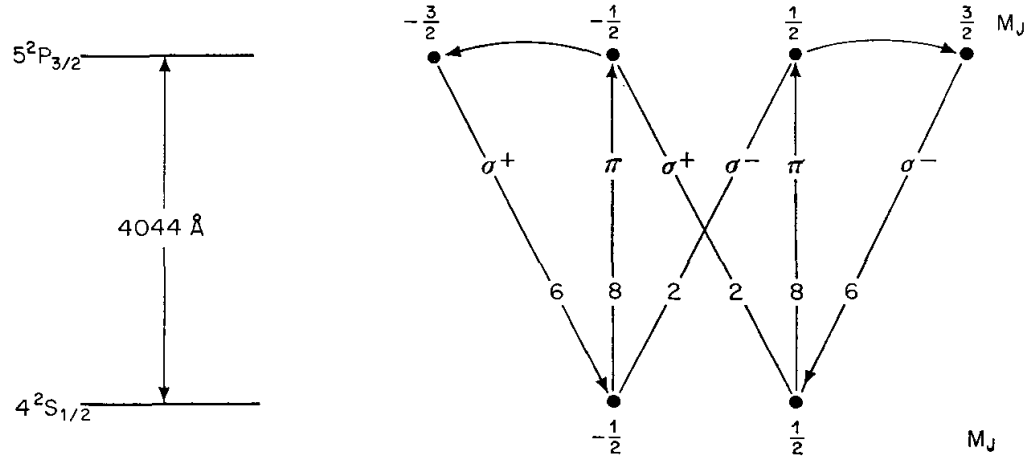


FIG. 6. Fine structure of the transition $4^2S_{1/2} - 5^2P_{3/2}$ showing Zeeman components and relative transition probabilities. (After Ritter and Series.[2])

With I and J decoupled, both the optical and the radio-frequency interactions are with the electron, leaving the orientation of the nucleus unchanged. The interactions, showing the changes in M_J, are indicated in Fig. 6. The resonances corresponding to transitions between the four values of M_J would

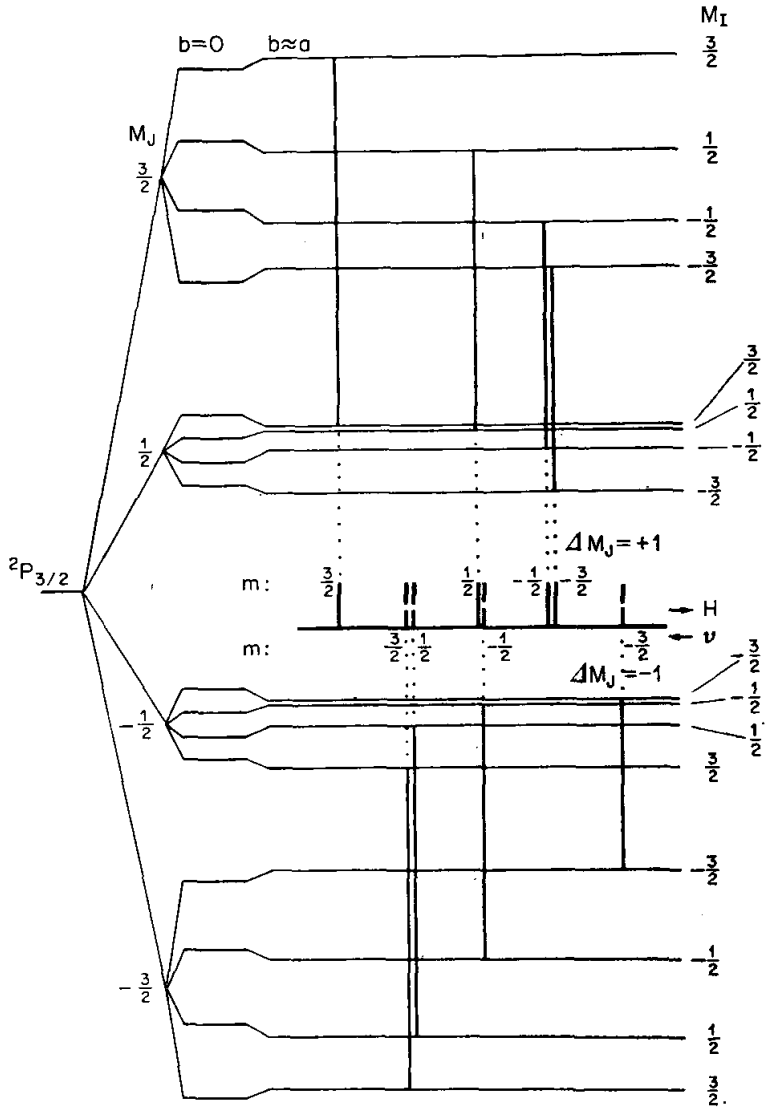


FIG. 7. Hyperfine structure of Zeeman components of $5^2P_{3/2}$ showing the radio-frequency transitions detectable by the experimental arrangement shown in Fig. 3. The figure has been drawn for a value of the electric quadrupole coupling constant b approximately equal to that of the magnetic dipole constant a. (After Ritter and Series.[2])

be coincident were it not for the hyperfine coupling which produces the splittings indicated in Fig. 7.

The corresponding resonance curve is shown in Fig. 8. The frequency was fixed at 41 MHz and the curve obtained by varying the magnetic field. A low-frequency modulation was added to the field in order to allow phase-sensitive

detection. The curve is therefore the derivative of the true resonance curve. The structure is hyperfine structure. Analysis of this curve allowed the determination of the hyperfine interaction constants—magnetic dipole and electric quadrupole.

A recent analysis (Pegg[3]) has shown that the quadrupole constant obtained from these curves was seriously in error owing to perturbation of the resonances by the strong radio-frequency fields. These perturbations will be discussed in a later section.

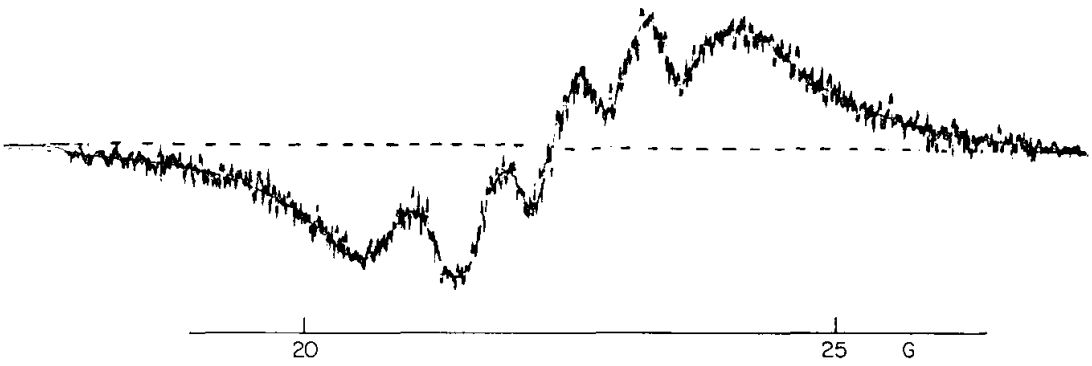


FIG. 8. Experimental resonance curve at 41 MHz (derivative curve). The structure represents the eight incompletely resolved transitions shown in Fig. 7. (After Ritter and Series.[2])

3.3. BACKGROUND SIGNAL

In Fig. 9 are compared two signals, one obtained with an oscillating field at 19 MHz and the other with this field turned off. In the first curve the magnetic resonance with hyperfine structure is seen, as in Fig. 8, but a background signal is present which the second curve shows to be independent of the radio-frequency field. There are two contributions to this background signal:

(i) It occurs in the region where the I–J coupling is being broken down. In this region the atomic wave functions are field-dependent, and the optical transition probabilities are also field-dependent. (In the early days of resonance fluorescence, attemps were made to determine nuclear spins by measuring these changes: Heydenburg *et al.*[4])

(ii) In this region (and also near zero field) there are degeneracies of energy levels. This gives rise to interference effects which are now known as level-crossing and anti-crossing effects. (The changes of intensity in Fig. 9 are due to anti-crossings because π radiation was used to excite the fluorescence). These interference effects form the basis of what has proved to be a most important spectroscopic technique. They were not recognised when the experiments on potassium were performed: the changes of intensity were ascribed to the effect (i). Recent studies (Schmieder *et al.*[5]) of level-

crossing effects in these hyperfine states of potassium have led to values of the coupling constants free from the error introduced by the strong radio-frequency fields.

The next sections will be devoted to these interference effects.

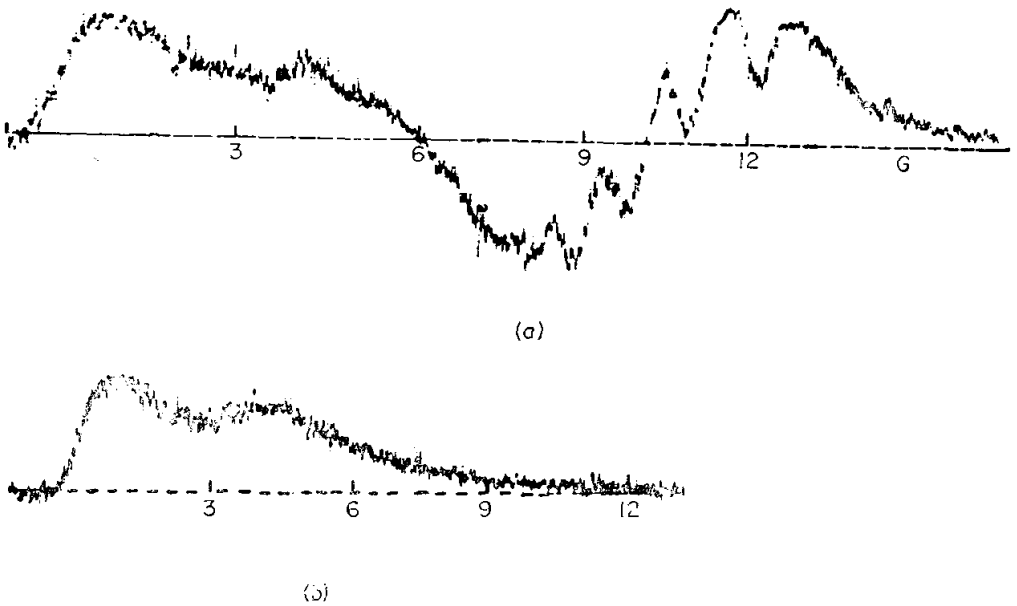


FIG. 9. Fluorescence signal in the range 0–15 G. (After Ritter and Series.[2])
a. With radio-frequency field at 19 MHz.
b. With no radio-frequency field.

4. LEVEL-CROSSINGS

4.1. THE PHENOMENON

First, I should like to show the phenomenon and indicate its usefulness, and then discuss its interpretation.

A sketch showing the essential parts of the apparatus, can be seen in Fig. 10. There is no radio-frequency field. Notice that the exciting light is linearly polarized perpendicular to the magnetic field, and that the fluorescent light (consisting in this case of two circularly polarized components emitted along the direction of H) is taken through a linear analyser before reaching the photomultiplier. I emphasise that, for the exciting light and for the fluorescent light we are using one mode of polarization (linear polarizers), but that two different atomic transitions are involved ($\Delta M = \pm 1$), both in excitation and in decay.

The experiment is performed by recording the intensity of fluorescent light as a function of magnetic field. Figures 11a and 11b show results for fluorescence from an excited state of thallium ($(6s^2 6d)^2 D_{3/2}$).[6] In this particular

experiment, low-frequency modulation of the magnetic field was used for phase-sensitive detection, so the figures show $\partial I/\partial H$.

In Fig. 11a you see strong changes of the intensity in the neighbourhood of zero field. In 11b you see changes at higher fields. These are superimposed on

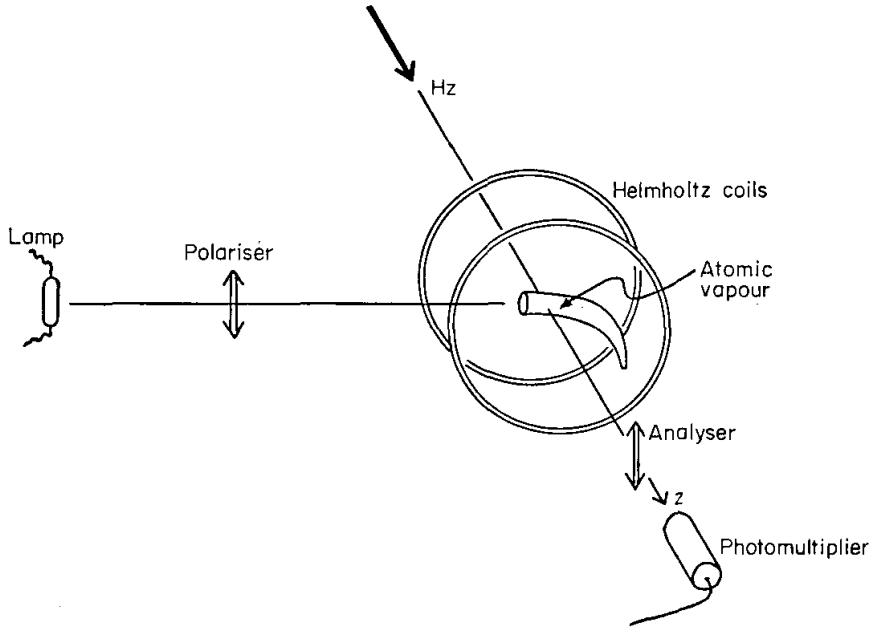


FIG. 10. Geometrical arrangement for studying the level-crossing phenomenon.

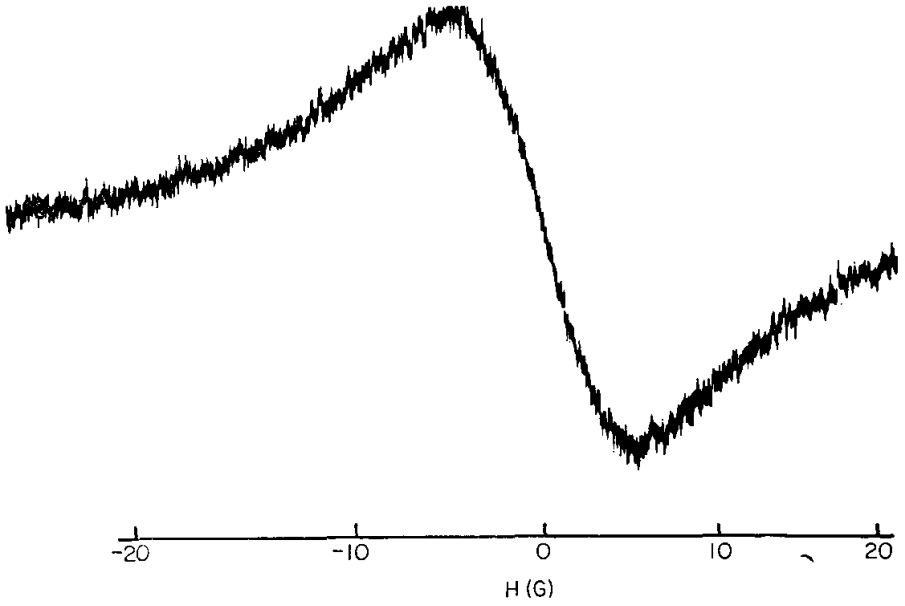


FIG. 11a. Resonance fluorescence from $(6s^2 6d)\ ^2D_{3/2}$ state of thallium in the vicinity of zero magnetic field. (After Gough and Series.[6])

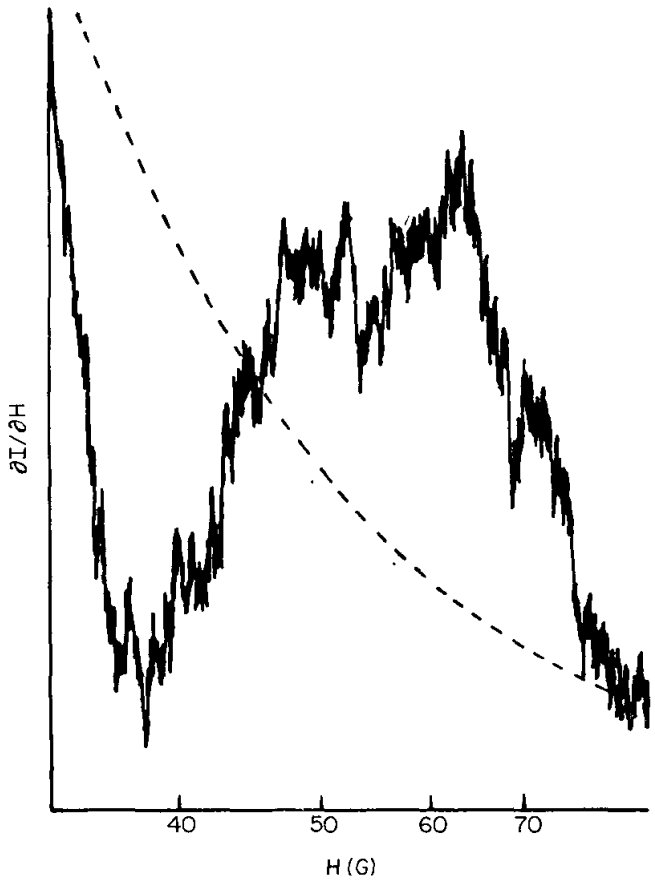


FIG. 11b. The signal extended to higher values of magnetic field. The sensitivity has been increased. The broken line shows the tail of the curve of Fig. 11a. (After Gough and Series.[6])

the tail of the zero-field curve which is shown by the broken line, and the gain has been substantially increased.

Now look at the energy level diagram, Fig. 12, and notice that energy levels having $\Delta M = 2$ intersect at zero field and at two other places in finite fields. The degeneracy of levels is held to be responsible for the changes in intensity of the fluorescent light. Experimentally, one measures the value of the magnetic field at the centre of the anomaly and calculates the hyperfine constants with the help of the Breit–Rabi formula.

Measurement of the line width is also important. This is generally done for the anomaly at zero field. One can easily calculate the mean lifetime of the excited state from the measured line width.

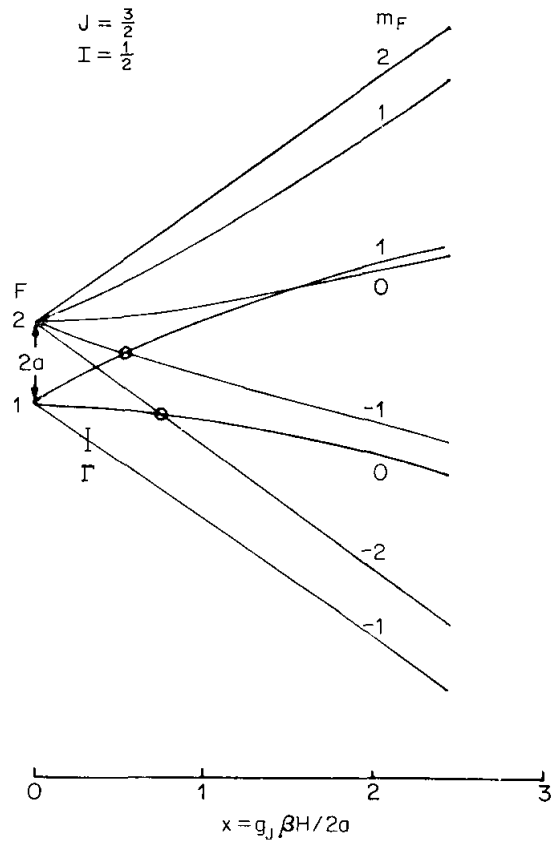


FIG. 12. Decoupling of hyperfine structure by the magnetic field. The vertical bar marked Γ represents the radiation width of the levels. a is the magnetic dipole constant. The signals of Fig. 11b are attributed to the intersections of levels marked with circles. (After Gough and Series.[6])

4.2. INTERPRETATION OF THE PHENOMENON

It is essentially an interference phenomenon. When the levels are degenerate and under the geometrical conditions specified above, the atoms are excited coherently in two modes and by virtue of this, can radiate coherently in two modes. The interference results in a redistribution in space of the fluorescent light, compared with the distribution when the modes are excited incoherently. The decay rate of the atoms is unaffected: there is no change in the intensity of the fluorescent radiation integrated over space. Notice that the redistribution of light is attributable here to interference whereas in the double resonance experiments it was attributable to the radio-frequency field.

The possibilities for interference are apparent from Fig. 13. The important points are: for the atoms—one initial state, one final state, two intermediate states; for the light—coherence in the incident beam and at the detector.

We can write simple equations based on the notion of pulse excitation which we used before. Suppose an atom is excited at time t_0 by a pulse of polarized

light which allows transitions from a common initial state $|g\rangle$ to either of two excited states $|\alpha\rangle$ or $|\beta\rangle$. The wave functions for $|\alpha\rangle$ and $|\beta\rangle$ develop according to $\exp -i(k_i - \frac{1}{2}i\Gamma_i)(t-t_0)$ $(i = \alpha, \beta)$, where k_i is the eigenfrequency and Γ_i the damping constant. In many applications (for example, in cases of hyperfine structure), we have $\Gamma_\alpha = \Gamma_\beta$. The state vector at time t for those atoms excited at t_0 is (to within a constant),

$$|t, t_0\rangle = \sum_{i=\alpha,\beta} P_{ig} \exp -i(k_i - \tfrac{1}{2}i\Gamma)\,(t-t_0)\,|i\rangle, \tag{4}$$

where P_{ig} is the (time-independent) matrix element of the electric dipole operator between the ground and excited state. (This is written as a plausible equation: it can be justified in a number of ways.) The intensity of the light emitted at time t in such a direction and polarization as to reach the detector will be proportional to $|\langle f|P'|t, t_0\rangle|^2$. Thus

$$\begin{aligned} I(t, t_0) \propto\ & [|P'_{f\alpha} P_{\alpha g}|^2 + |P'_{f\beta} P_{\beta g}|^2] \exp -\Gamma(t-t_0) \\ & + P'_{f\alpha} P_{\alpha g} P'_{\beta f} P_{g\beta} \exp -i(\delta - i\Gamma)(t-t_0) \\ & + \text{complex conjugate}, \end{aligned} \tag{5}$$

where $\delta = k_\alpha - k_\beta$.

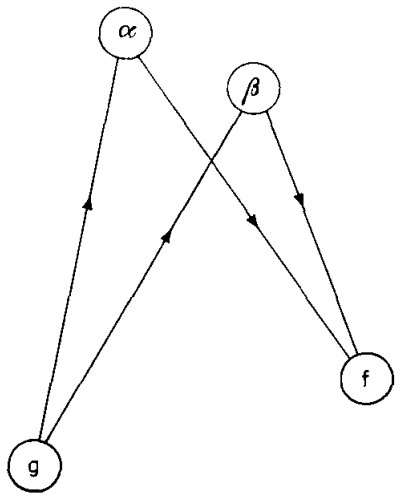


FIG. 13. Resonance fluorescence by two paths from initial state g to final state f.

In a steady-state experiment we integrate, as before, over t_0 from 0 to t and obtain

$$I \propto \frac{(C_1^2 + C_2^2)}{\Gamma} + A\frac{\Gamma}{\Gamma^2+\delta^2} + B\frac{\delta}{\Gamma^2+\delta^2} \tag{6}$$

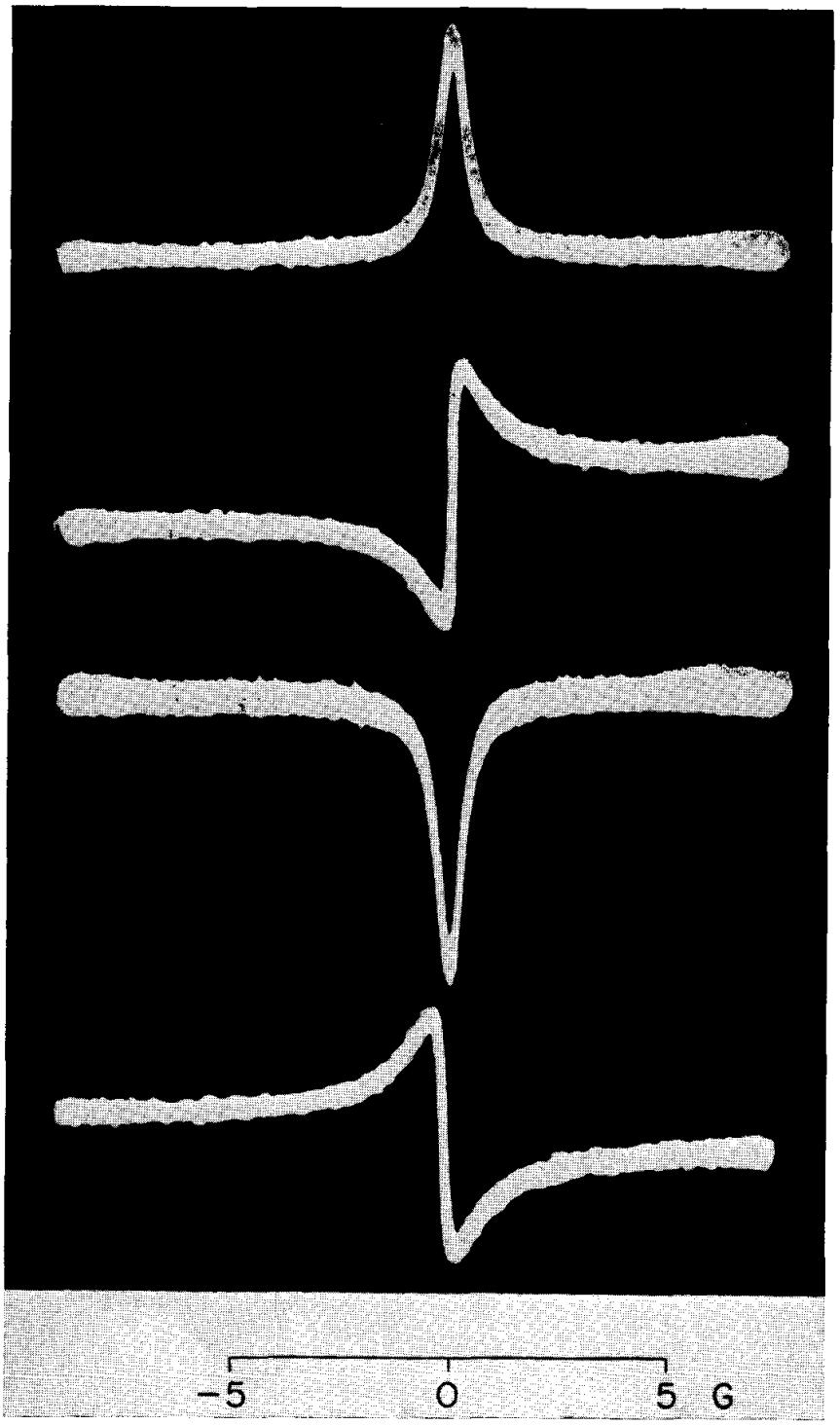


FIG. 14. Zero field level-crossing signals (Hanle effect) for 6^3P_1 in mercury. The four curves correspond to rotation of a polarizer in successive steps of $\pi/4$. (After Kibble and Series.[7])

where $C_1 = |P'_{f\alpha} P_{\alpha g}|$, $C_2 = |P'_{f\beta} P_{\beta g}|$, and $A + iB = P'_{f\alpha} P_{\alpha g} P'_{\beta f} P_{g\beta}$. (7)

The first term of (6) is a constant. The second and third are Lorentzian bell-shaped and dispersion functions of δ. These are the interference terms, which vanish when $\delta \gg \Gamma$. By suitable orientation of polarizer and analyzer one can make either A or B vanish. These results are illustrated in Fig. 14,[7] which shows a set of experimental curves obtained for the zero-field level-crossing in 6^3P_1 states in mercury—the states studied in the Brossel–Bitter experiment.

4.3. DETERMINATION OF LIFETIME

The width at half intensity (the bell-shaped curve) and the distance between the peak and the dip (dispersion curve) is equal to 2Γ. For the mean lifetime, τ, we have

$$\tau(\text{sec}) = \Gamma^{-1}(\text{radians per sec})^{-1} \tag{8}$$

The method of studying the zero-field level-crossings is, in principle, very attractive for the determination of lifetimes because the sole perturbation on the atoms is the incident light, which is a negligible effect in comparison with spontaneous emission. But in practice there are a number of difficulties:

(i) admixture of the bell-shaped and dispersion curves,

(ii) multiple scattering of resonance radiation. This leads to an anomalous narrowing of the curves called "coherence narrowing".

(iii) distortions caused by non-uniform spectral distribution of the incident light.

We shall discuss these problems in a later section. In practice, the method can be applied to measure lifetimes to a precision of 1 % in favourable cases.

5. ANTICROSSINGS

There is nothing in the above analysis which requires the levels to be of different M—the interference terms still exist if the levels have the same value of M. But, in time-independent perturbation theory, "levels of the same M never cross". If we are considering the breakdown of I–J coupling, for example, the hyperfine interaction $a\mathbf{I}.\mathbf{J}$, which has non-vanishing matrix elements between states of the same value of $M = (M_I + M_J)$, causes repulsion of such levels. Nevertheless, a pair may approach one another to within their natural width, and in this case the interference effect is not negligible. Such a situation is called an "anticrossing".[8]

In the region of an anticrossing the wave functions of the perturbing states are changing rapidly. For this reason it is easier to base an analysis on the unperturbed states, say $|a\rangle$ and $|b\rangle$. The optical matrix elements to $|a\rangle$ and $|b\rangle$ vary only slowly through the anticrossing. Calculation of the change in

intensity of the fluorescent light shows that the interference effect has its greatest value when either $|a\rangle$ or $|b\rangle$ alone are populated. The interference signal has the analytical form $|2V|^2/(\gamma^2+\delta^2+|2V|^2)$, where $|2V|$ is the interval between the levels at their closest approach (a measure of the perturbation energy). This expression should be compared with eqn (3) which describes a double resonance curve. The two have exactly the same form. An anticrossing signal is the same as a double resonance signal, but in the one case the perturbation is internal ($|2V|$), and in the other, external (γH_1). (See Fig. 15)

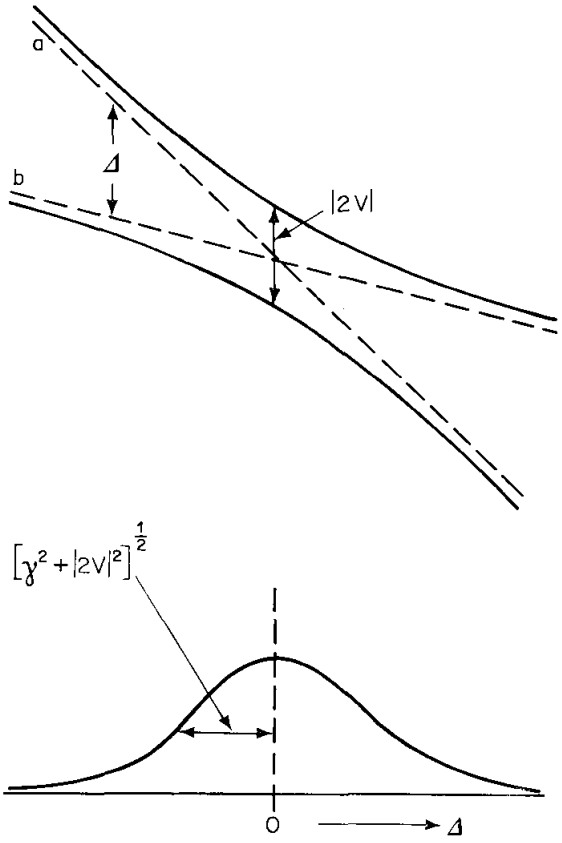


FIG. 15. An "anti-crossing" of energy levels and the corresponding fluorescence signal whose analytical form is given in the text. Here, γ is the damping constant.

It is to be noticed that anticrossing signals are broader than crossing signals, and are not suitable for the measurement of lifetimes. On the other hand, they can be analysed to yield the hyperfine interaction constants.

6. GROUND STATES: OPTICAL PUMPING

We have described methods which allow the study of excited states of atoms. To complete our preliminary review we describe some typical experiments in which the ground states of atoms are the focus of interest. Therefore we must choose as a suitable substance one whose atoms have structure in the ground

states. (The even isotopes of mercury, which served as an example before, have a ground state without structure.)

The alkalis have the required structure. Rubidium serves as an excellent example because its vapour pressure at ordinary room temperatures is about right for the experiments we shall describe.

6.1. THE PUMPING PROCESS

Consider an assembly of atoms which have undergone resonance fluorescence by anisotropic excitation followed by spontaneous emission, which is an isotropic perturbation. In principle, we have injected anisotropy into the system; that is, we have altered the distribution of atoms over the ground states. We have, therefore, prepared the system for magnetic resonance experiments.

Points of detail will emerge by considering a simple experiment. (See Fig. 16)

The bulb contains rubidium vapour at a pressure of about 10^{-6} torr. (It

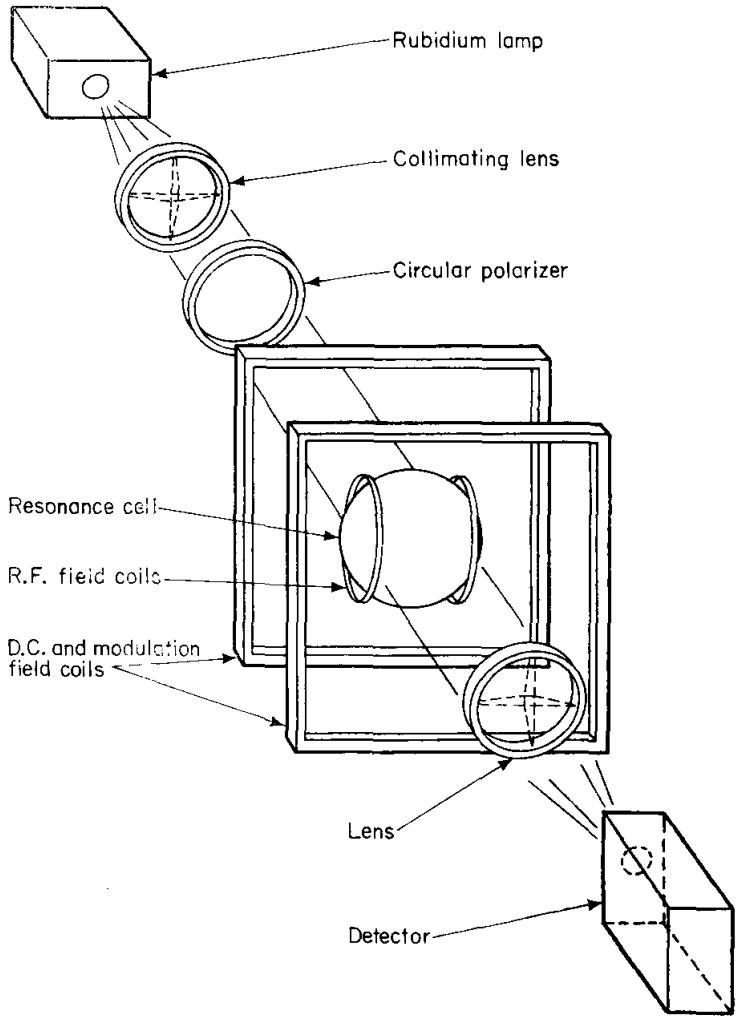


FIG. 16. Apparatus for studying optical pumping in rubidium.

also contains a noble gas at a pressure of a few torr for a reason we shall shortly explain). It is illuminated with light from a rubidium lamp which passes through a circular polarizer. The mounting allows a magnetic field to be applied in the direction of the incident light, and also enables this direction to be aligned with the Earth's field, since this is by no means to be ignored.

The optical transitions are indicated in Figs. 17a and b which, for the sake of simplicity, ignore hyperfine structure. It is clear that, if the atoms are excited by σ^+ light and decay by spontaneous emission without any other perturbation, the net result of one cycle will be to transfer atoms from the ground

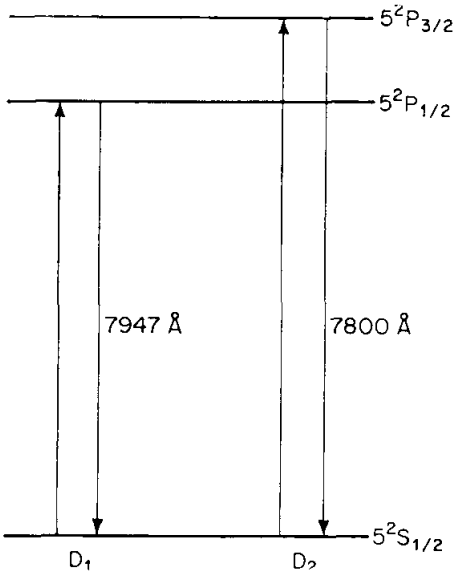


FIG. 17a. Resonance fluorescence of the principal doublet in rubidium.

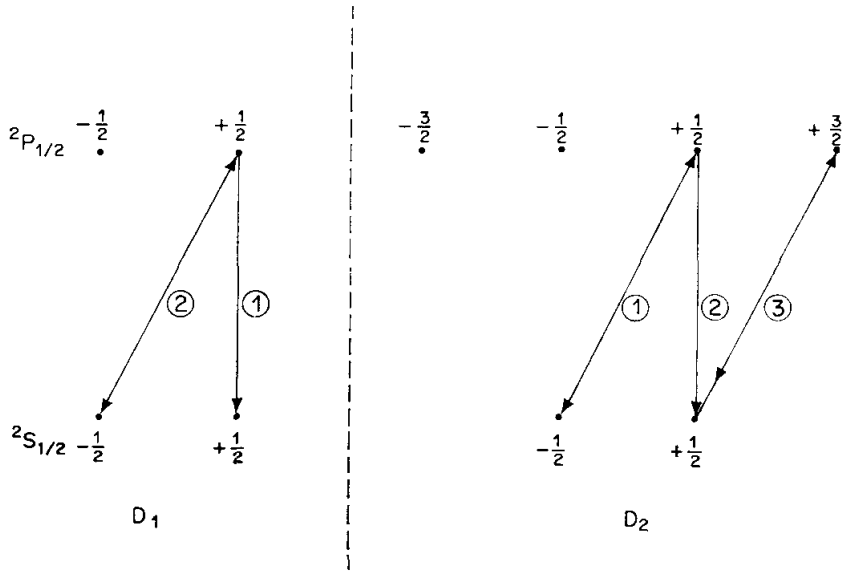


FIG. 17b. Transitions induced by excitation with circularly polarized light. The numbers indicate relative transition probabilities.

state $|-\frac{1}{2}\rangle$ into $|+\frac{1}{2}\rangle$. This may be expressed quantitatively. Starting from equal populations of the two states, the D_1 light alone will produce a 20:40 mixture, while the D_2 light alone will produce a 10:50 mixture. The two effects are additive. (We shall reconsider this point when we investigate the effect of the noble gas.)

6.2. RADIO-FREQUENCY RESONANCE

Having established a difference of population, we can now contemplate a resonance experiment. Coils, as shown in Fig. 16, provide an oscillating field at right angles to the direction of the light beam. The required field amplitude is only of the order of milligauss, since the atoms have lifetimes of the order of milliseconds (we discuss this below). The frequency may be of the order of kHz if we are studying transitions between Zeeman components, or of the order of 10 to 10^3 MHz if we are studying hyperfine transitions. Signal generators will frequently provide enough power.

6.3 MONITORING THE RESONANCES

The r.f. field alters the populations of the levels, and this may be detected

(i) by its effect on the fluorescent light (but this method will not be applicable if the bulb contains a gas), or

(ii) by its effect on the absorption coefficient of the vapour.

This latter method has been most commonly used, and is illustrated in Fig. 16. The same light which does the pumping also monitors the resonance. Reference to Fig. 17 shows that the amount of circularly polarized light absorbed depends on the distribution of atoms between the ground states. The resonance is indicated by changes in the current of the photocell placed behind the resonance vessel.

6.4. DISCUSSION OF THE RESONANCES

Fig. 18 is an oscilloscope trace of resonances obtained with apparatus of this sort. It is rather a crude picture, but it brings out important points. Notice the following:

(i) stray fields have been compensated to about 2 mG. The only remaining field is the longitudinal field shown on the scale;

(ii) R.f. frequency: 5 kHz. Amplitude: a few mG;

(iii) there is a peak at zero field which needs to be explained;

(iv) resonances at $\pm 10{\cdot}7$ mG are due to ^{85}Rb, at $\pm 7{\cdot}0$ mG due to ^{87}Rb; (natural abundances 72%, 28%; nuclear spins 5/2, 3/2; g_F, 1/3, 1/2; respectively);

(v) resonance line-widths are ~ 2 mG (^{85}Rb) ~ 1 kHz, so that $\tau \sim 10^{-4}$ sec (assuming the line is homogeneously broadened);

(vi) the small wiggles are due to pick-up from the mains, showing that the time for traversing the sweep was of the order of seconds. This is necessary in order to avoid distorting the resonances, and emphasises the relatively large value of τ.

Consider point (iii). Near zero field the energy levels are degenerate or near-degenerate. There are stray transverse static fields and also transverse, low-frequency oscillating fields from the electrical mains. These can stimulate a resonance at zero frequency (or 50 Hz). These stray fields can account for the peak at zero field.

Some conceptual difficulty may still remain. While it is easy to use quantum labels M_F for non-degenerate levels, and to understand transitions between them, can one still use such language when the levels are degenerate? The answer is yes, for one can choose a quantum axis arbitrarily, and describe any experiment thereby, provided one takes proper account of the perturbations. Here, we choose an axis along the light beam and describe the optical transitions with reference to that axis. We notice that the magnetic field may be skew to the axis and we use a formalism which takes account of that, namely, magnetic resonance transitions at zero frequency. We could equally well

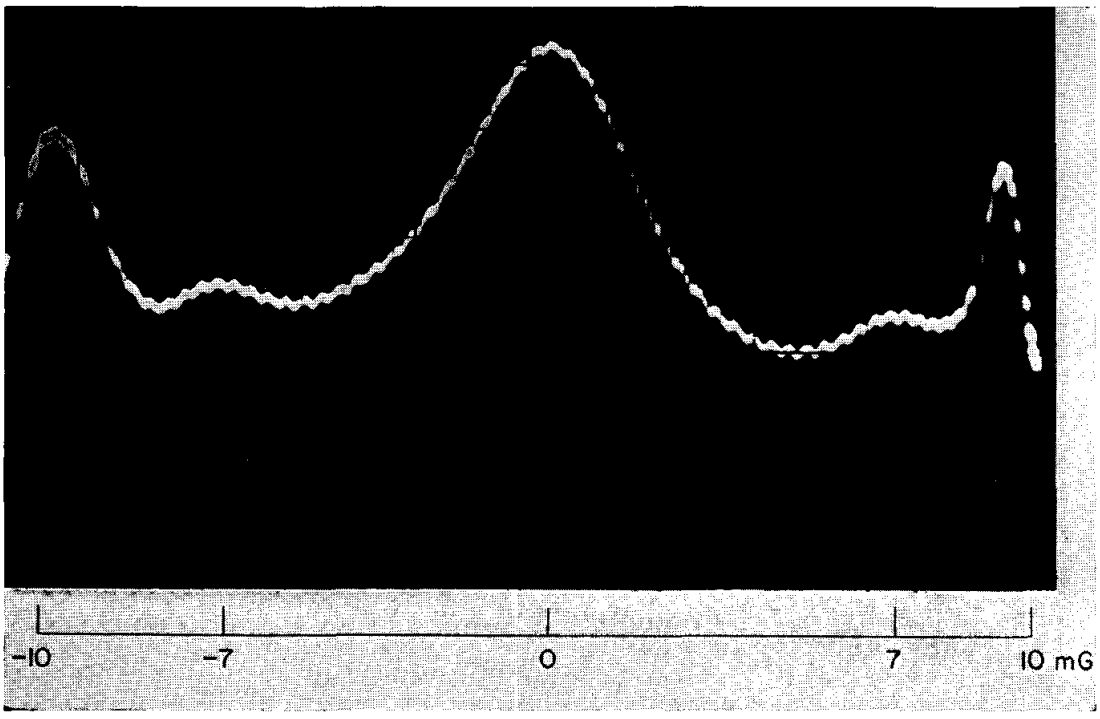


FIG. 18. Oscillogram demonstrating radio-frequency resonances in the ground state of rubidium studied by optical pumping. Resonances at ± 7mG correspond to ^{87}Rb; those at ± 10.7mG to ^{85}Rb; for other features see text.

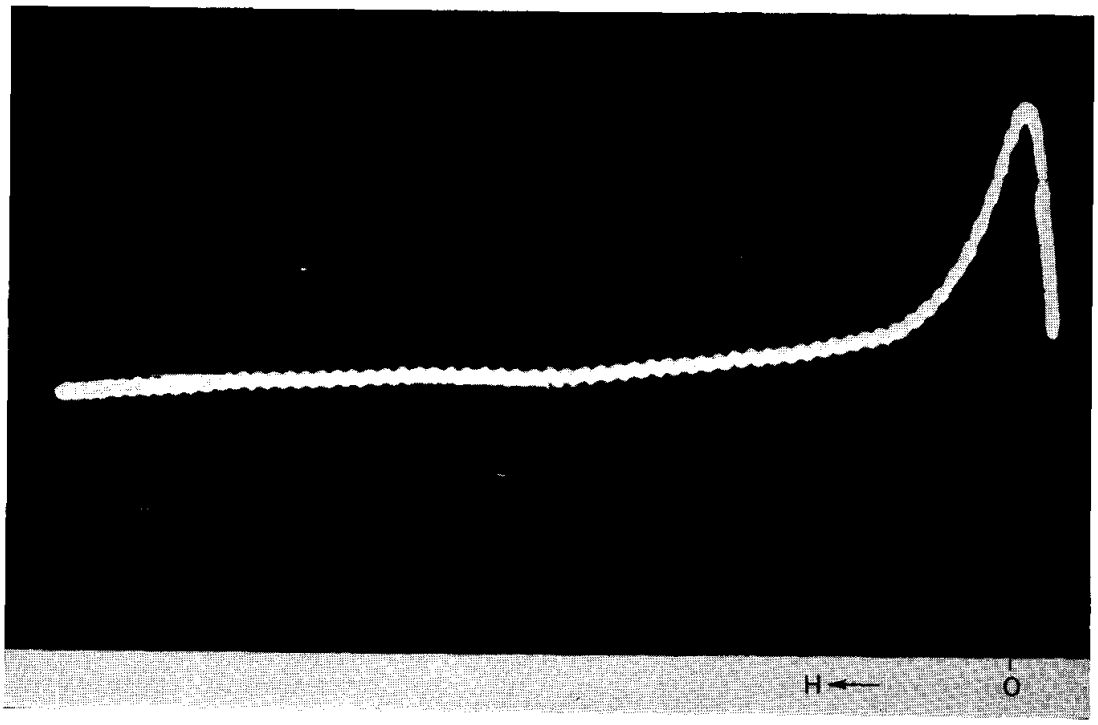


FIG 19a. Optical pumping signal in rubidium. No r-f field.

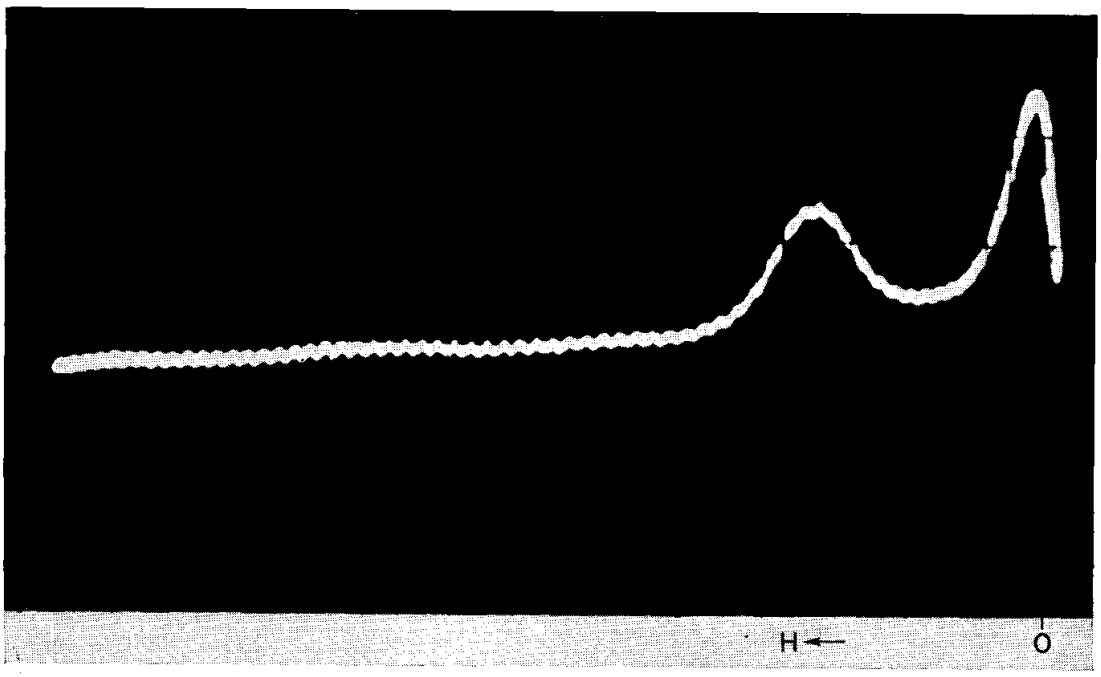
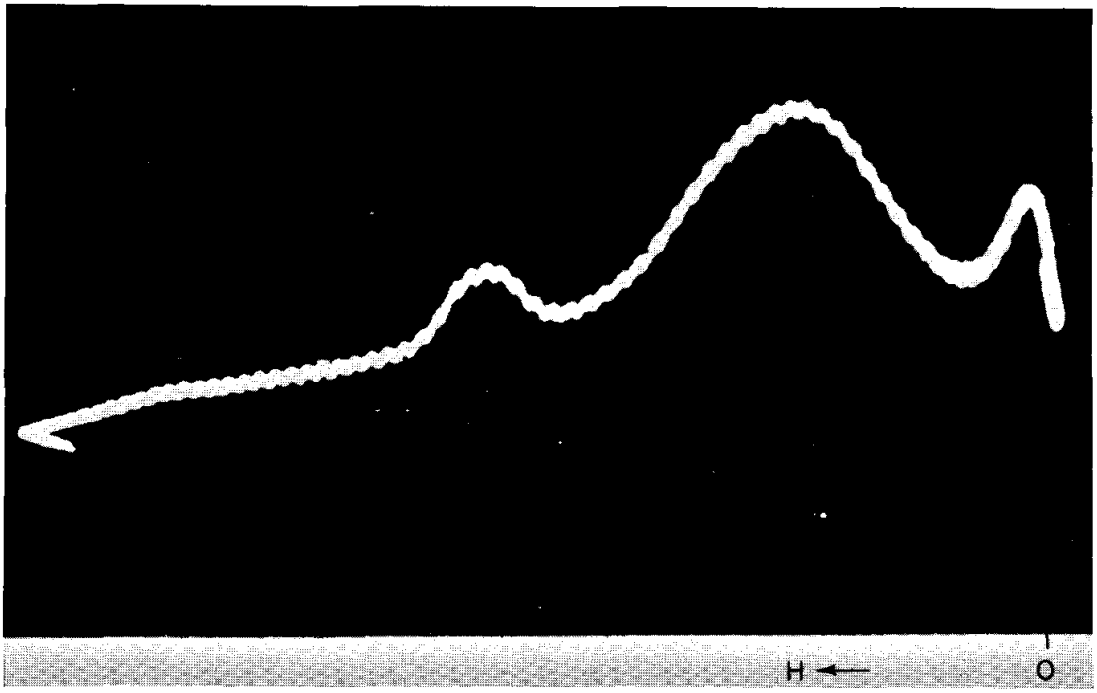


Fig. 19b. Resonance at a frequency of 3 kHz.



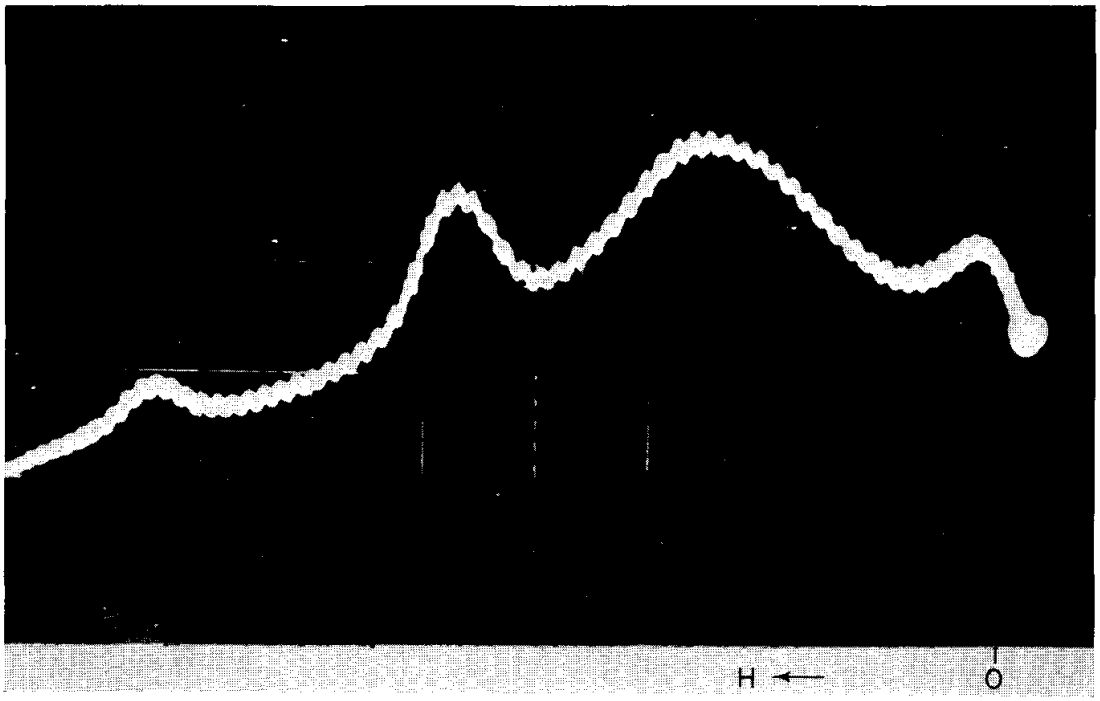


FIG. 19 c, d. The effect of increasing r-f power: resonances at multiples of the field H_0.

describe these transitions by writing equations for rotating the axis from the longitudinal to the skew direction. Alternatively, we might choose an axis in the skew direction and describe the optical transitions with reference to such an axis, but we would have to remember that, on account of the degeneracy, there may be coherence between different optical transitions.

The fields of work opened by resonances such as shown in Fig. 18 are:

(a) precision measurement of g-values of atoms in ground states, (and, correspondingly, of hyperfine intervals);

(b) measurement of nuclear spins. (The values of g_F may be determined from the measurements. Then, if g_J is known, I may be deduced);

(c) precision magnetometry and atomic clocks;

(d) studies of relaxation processes by measurement of the line width under different conditions. We shall elaborate on this.

6.5. MULTIPLE QUANTUM RESONANCES

We now illustrate in Fig. 19 an effect which can arise very easily in ground state optical pumping experiments, and can lead to mistaken identification of resonances. This is the occurrence of what appear to be harmonic resonances.

The magnetic field sweep is the same as in Fig. 18, but the zero has been displaced to the right. The radio-frequency has been reduced from 5 to 3 kHz. The figures a, b, c, d are taken with increasing r.f. power, starting from zero. Curve (a) is the zero-field resonance. In (b) is seen the resonance of ^{85}Rb, as in Fig. 18 but at a field H_0 about 6·4 mG. The resonance of ^{87}Rb is unresolved.

Increasing r.f. power, Figs. 19 (c) and (d), brings up resonances at approximately $2H_0$ and $3H_0$. (They are not exactly at integral multiples of H_0, but the small displacements are not our present concern). These might have been due to harmonics in the oscillator, but this possibility was excluded. They are

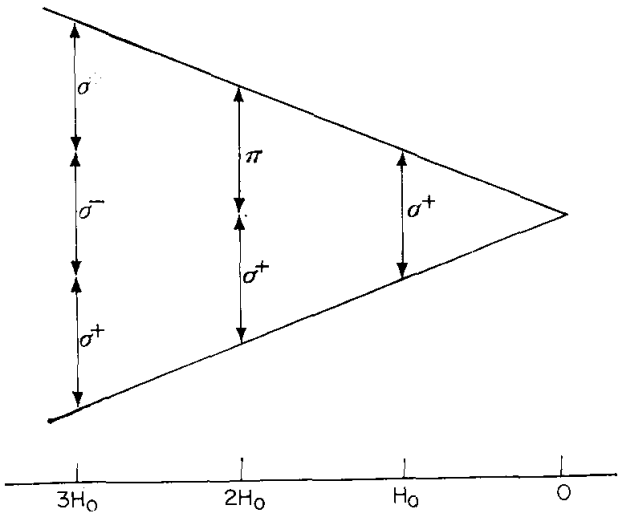


FIG. 20. The resonances interpreted as multiple quantum transitions.

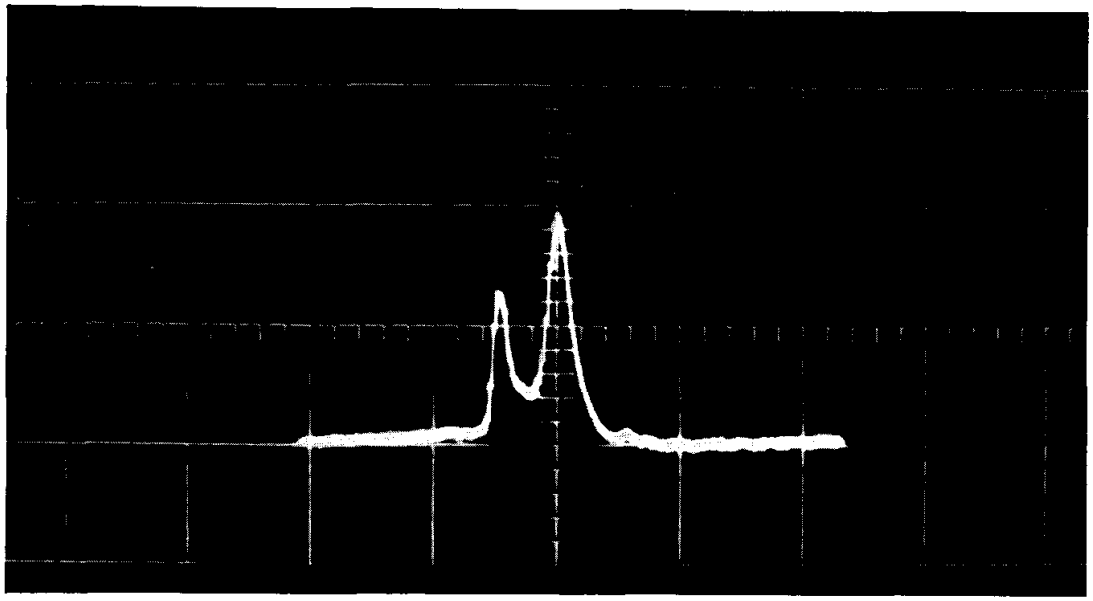


FIG. 21a. Resonance at 5 kHz produced by rotating field. (The zero field signal is also present).

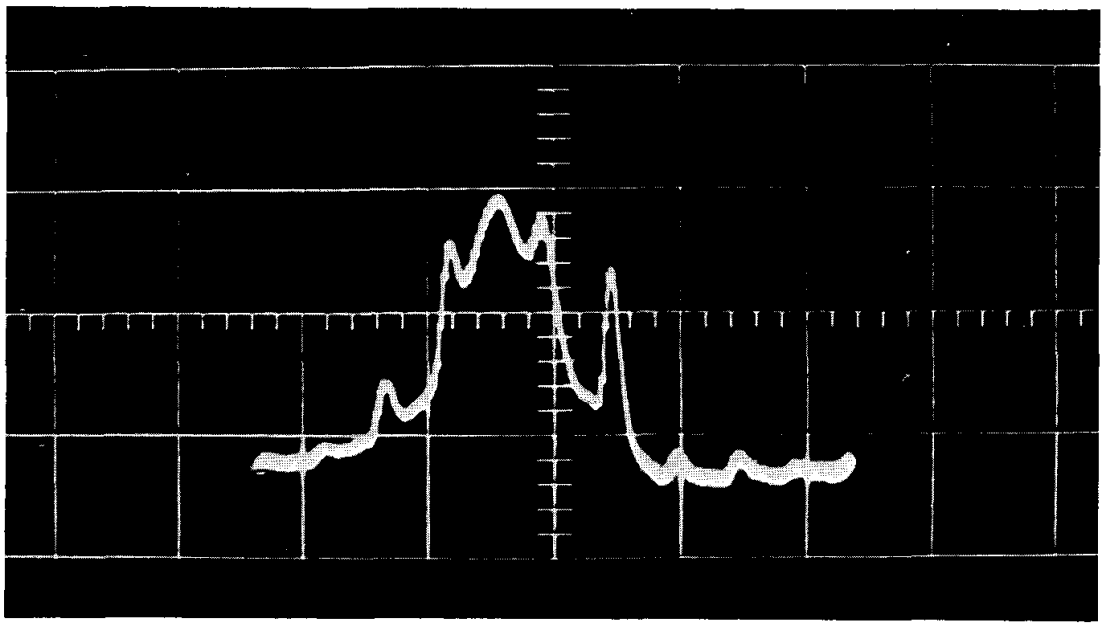


FIG. 21b. Effect of increasing r-f power. The dominant resonance is undisplaced but greatly broadened. Numerous subsidiary resonances appear; the zero field signal is displaced slightly to the left (see Fig. 42).

interpreted as multiple quantum transitions between a pair of levels having $\Delta M = \pm 1$, and require for their interpretation the existence of components of the r.f. field *other than* the rotating field which is responsible for the primary resonance at $H_0(\sigma^+ : \Delta M = \pm 1$ in absorption).

Figure 20 shows how a component of the oscillating field parallel to the static field (π: $\Delta M = 0$), combined with $\sigma^+(\Delta M = \pm 1)$, can satisfy the conservation of energy and of angular momentum at a field $2H_0$, while the counter-rotating component $\sigma^-(\Delta M = -1$ in absorption) can combine with $2\sigma^+$ to satisfy the conservation laws at $3H_0$.

In Fig. 21a. is a resonance curve taken with an arrangement of coils designed to produce a rotating field (σ^+ alone). The frequency is now 5 kHz and zero field has been shifted to the centre. The zero field resonance is seen as well as the resonance at about 10 mG due to the rotating field. This should be compared with Fig. 18 where the resonances appear on both sides of zero because an oscillating field was used.

In Fig. 21b. the r.f. field strength has been greatly increased. A variety of resonances appear at fields up to $5H_0$ on the left and $4H_0$ on the right. These are due to small σ^- and π components arising from the non-uniform field of the coils, and acting with the σ^+ component to generate the resonances.

These multiple quantum transitions should not be confused with the multiple quantum transitions described above (Section 3.1 and Fig. 4b). In the present case we have transitions between atomic eigenstates induced by different components of the r.f. fields, acting coherently. In the former case we had transitions via non-resonant intermediate states of the atom induced by one particular component of the field.

These multiple quantum transitions have been studied by a number of authors.[9, 10]

6.6. WIDTH OF RESONANCE CURVES: RELAXATION TIMES

We have been concerned with the *occurrence* of resonances. We now return to the question of their *width.*

One might expect the width to be determined by the inverse of the time of flight of atoms across a resonance cell ($\sim 10^{-4}$sec, for example). In fact, resonance curves are frequently studied whose width corresponds to times of the order of 1 sec. This is generally achieved by using *wall coatings* or *buffer gases.* The important fact is that, in many cases of interest, it is possible for atoms to undergo many thousands of collisions either with gas molecules, or with the coated wall, without losing their orientation. This is generally true for atoms whose ground states are S states (such as the alkalis). Their orientation arises from the electron and nuclear spin, but the spherical symmetry of the distribution of charge isolates the spins from the effects of collision.

Substances chosen for wall coatings are chemically inert materials such as

paraffin hydrocarbons. Similarly, noble gases at pressures from a few torr up to atmospheres are used for buffer gases.

6.7. PUMPING CYCLE WITH BUFFER GASES

The use of buffer gases has important consequences for the mechanism of the pumping process, since excited states (which will be P states if the ground states are S) are not inert under collisions with buffer gases. When the pressure of the gas is raised to the point where the time between disorienting collisions is comparable with the radiative lifetime of the pumped atoms, the changes of population induced by optical pumping are radically changed. The situation is usually analysed by assuming that collisions result in the random orientation of excited atoms—equal population of the states $|M_F\rangle$ or $|M_J\rangle$. This is an oversimplification, but it gives useful qualitative results.

If all excited states are equally populated and decay at the same rate, then all the ground states are replenished at the same rate (whatever the coupling scheme). It follows that differences of population between ground states can only be established through different rates of excitation and this raises a difficulty when the ground states are S states. The intensity sum rules guarantee that if the absorption from an S state is summed over a multiplet (e.g. $S_{1/2}$ to $P_{1/2}$ and $P_{3/2}$), the transition rates from all components of the ground state are equal. (This may be verified by adding the transition probabilities in Fig. 17). Therefore, in order to achieve differences of population one must have light whose spectral distribution over the fine structure components is non-uniform. This is commonly achieved by the use of interference filters or absorption cells. Under some conditions the required change of spectral profile occurs as the pumping light traverses the vapour in the resonance vessel.

6.8. HYPERFINE PUMPING

Hyperfine pumping is another variant of the optical pumping method which relies on non-uniform spectral distribution of the pumping light. Suppose the hyperfine components in an absorption line are optically resolvable (resonance lines of rubidium, for example), it is then sometimes possible to find a lamp (often an isotope of the same element) whose spectrum covers the hyperfine components unequally (^{85}Rb (lamp) and ^{87}Rb (sample)). In such a case, light of any polarization will produce an inequality of population between hyperfine states of the ground level. The hyperfine intervals in zero field can now be studied by magnetic resonance, just as for the excited states. The relaxation mechanisms for hyperfine transitions may be different from those for Zeeman transitions. The intensities of the resonances are a measure of $\langle \mathbf{I}.\mathbf{J} \rangle$.

6.9. SPIN EXCHANGE

We have described how atoms can be oriented by light. An important exten-

sion of the technique due to Dehmelt was the discovery that atoms of one species can be oriented through collisions with atoms of another species which themselves have been oriented by light. Thus, for example, with a mixture of sodium and rubidium vapours in a vessel, the sodium can be oriented by illuminating the bulb with rubidium light. Moreover, r.f. resonances in the sodium may be monitored with the rubidium light because the sodium atoms which have interacted with the radio-frequency field themselves interact with the rubidium atoms.

An important application of this spin-exchange technique is in the study of substances which cannot themselves be oriented by light, or for which there are technical difficulties. Among the substances so studied are atomic hydrogen (and its isotopes) and free electrons.

6.10. METASTABILITY EXCHANGE

Closely related to spin-exchange is metastability-exchange, first studied by Colegrove *et al.*[11] We shall illustrate this and the following sections by reference to the experiments of Greenhow.[12]

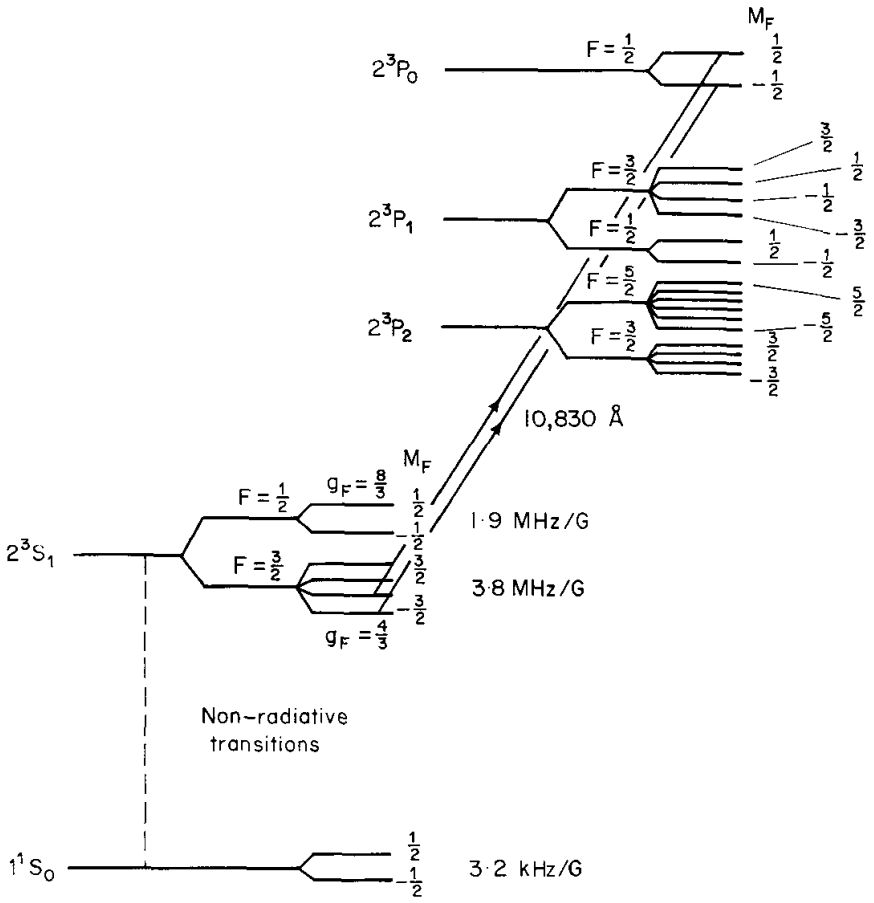


FIG. 22. Term diagram of ^{3}He showing optical pumping from the metastable state 2^3S_1.

Optical pumping is performed from the metastable state 2^3S_1 which is populated (with random orientation) in a discharge in helium gas at a pressure of a few torr (see Fig. 22). Illumination with circularly polarized light at 10,830 Å or 3889 Å from a helium lamp produces polarization of the metastable atoms through the intermediacy of the excited states 2^3P or 3^3P respectively. The polarization is reflected in the population of the hyperfine states of 2^3S_1, and if the I–J coupling is not broken down by an external field, the nuclei will also be polarized. When these metastable atoms collide with the much more numerous atoms in the ground state, 1^1S_0, there is a probability that the excitation energy will be transferred without affecting the nuclear polarization. Such collisions establish a nuclear polarization in the ground state.

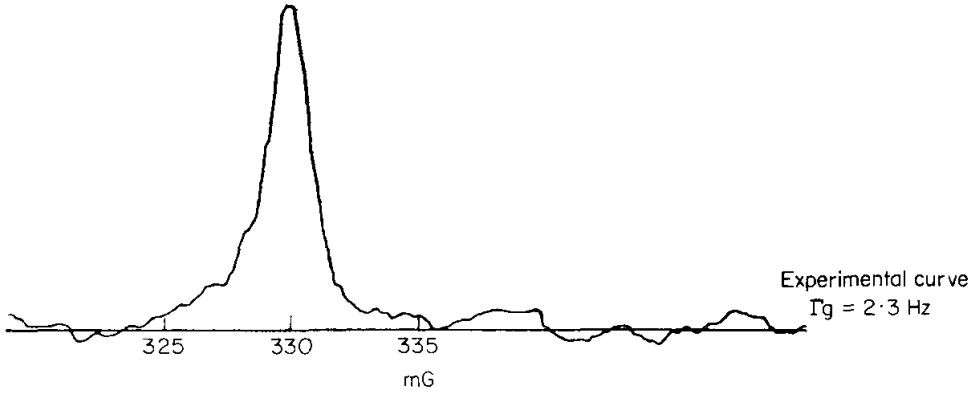


FIG. 23a. Nuclear resonance at a frequency of 1 kHz in the ground state of ^{3}He (this curve was taken with modulated light: see Section III 6.3).
(After Partridge and Series. *Proc. phys. Soc.* **88**, 983 (1966).)

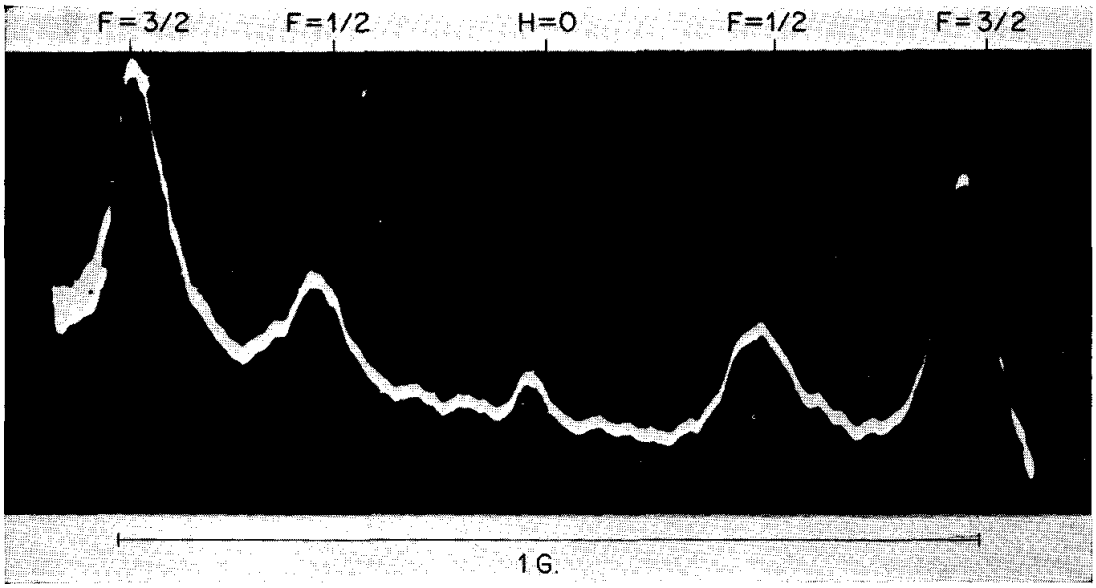


FIG. 23b. Resonances at 1 MHz in the metastable state. It is to be noticed that the line width is nearly two orders of magnitude greater than in (a).

It should be appreciated that this nuclear polarization is in a state in which the electronic angular momentum is zero: the magnetic moment of the atoms resides solely in the nuclear magnetisation. (For the metastable states, the electronic angular momentum is non-zero).

^{3}He atoms in their ground states have extremely long relaxation times. (Ten hours has recently been measured by Cohen-Tannoudji *et al.*[13]). In the gas discharge, relaxation times are of the order of minutes. The resonances and the relaxation times can be studied by measuring the absorption of light by the metastable atoms, though these are not the only methods available.

Figure 23a shows a nuclear resonance in ^{3}He at a frequency of 1 kHz. (Actually, this curve was taken with modulated light, but it demonstrates the characteristics of the resonance). The g-factor, about $2{\cdot}3 \times 10^{-3}$, shows that it is a *nuclear* resonance.

For comparison, Fig. 23b shows a set of resonances, also monitored by absorption of light by the metastable atoms, where the applied frequency is 1 MHz. The resonances occur at values of the field about $\frac{1}{4}$ and $\frac{1}{2}$ G respectively, whereas that in Fig. 23a is at about $\frac{1}{3}$ G. Yet the frequencies in the two cases differ by a factor 10^3. The resonances in (23b) are Zeeman resonances in the hyperfine states $F = 1/2$ and $3/2$ of 2^3S_1, where the g-factors, 8/3 and 4/3, are of electronic magnitude.

7. Relaxation Processes

In many applications of optical pumping it is of interest to study relaxation processes, for this gives information about the interactions between the optically pumped material and the collision partners. Methods available are static methods (e.g., measurement of resonance line width) and the study of transients. The number of time constants required to interpret the measurements depends on the complexity of the system and the symmetry of the environment. For a spin-$\frac{1}{2}$ system in an anisotropic environment two times are sufficient: the longitudinal (T_1) and transverse (T_2) relaxation times which are familiar from the Bloch equations of magnetic resonance.

The relaxation times will often be measured in a situation where the optical pumping process itself contributes to the relaxation: atoms are suddenly perturbed, either by optical excitation, or by collisions, according to the mechanism of pumping. The time constants measured will then be functions of the mechanism. These times we call τ_1 and τ_2. They are related to T_1 and T_2 by equations of the form

$$\frac{1}{\tau_1} = \frac{1}{T_1} + \frac{K_\alpha}{T_\alpha} + \dots$$

$$\frac{1}{\tau_2} = \frac{1}{T_2} + \frac{K_\alpha'}{T_\alpha} + \dots$$

where $T_\alpha \ldots$ are time constants for the pumping mechanisms, and K_α, $K_\alpha' \ldots$ are numerical constants depending on these mechanisms.

We give examples of the measurement of τ_1, T_1 and τ_2.

7.1 RELAXATION WITH THE PUMPING LIGHT: τ_1

Interpose a shutter between the pumping lamp and the cell. Suddenly withdraw the shutter and record the exponential change of intensity towards its equilibrium value (Fig. 24a). The time constant is τ_1.

7.2. RELAXATION IN THE DARK: T_1

When equilibrium has been reached, interpose the shutter for a definite time $(t_1 - t_0)$. During this time the sample has relaxed in the dark with time constant T_1. On removal of the shutter one obtains again the pumping curve (τ_1), but the initial point gives the polarization after the lapse of time $(t_1 - t_0)$. From the locus of these points (Fig. 24b) one obtains T_1.[14]

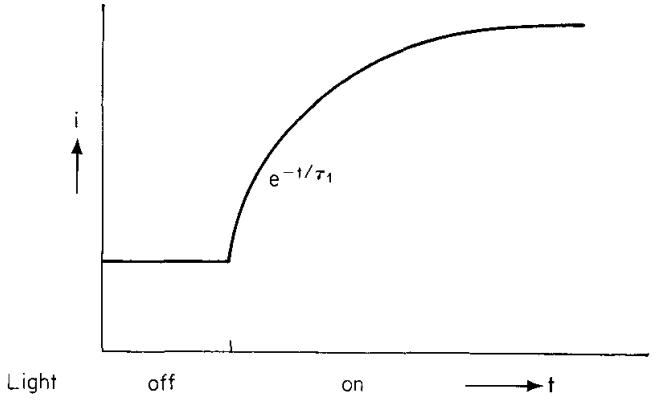


FIG. 24a. Exponential growth of optical pumping signal. (Time constant τ_1).

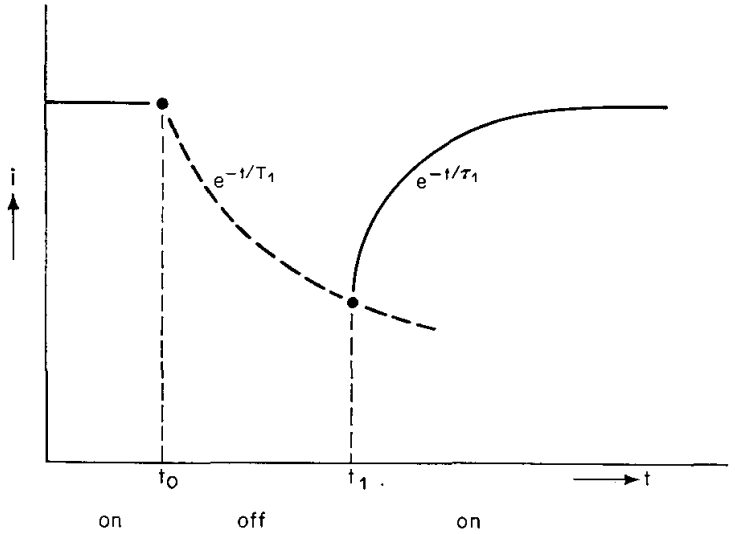


FIG. 24b. To illustrate the method of obtaining points on the curve for relaxation in the dark. (Time constant T_1).

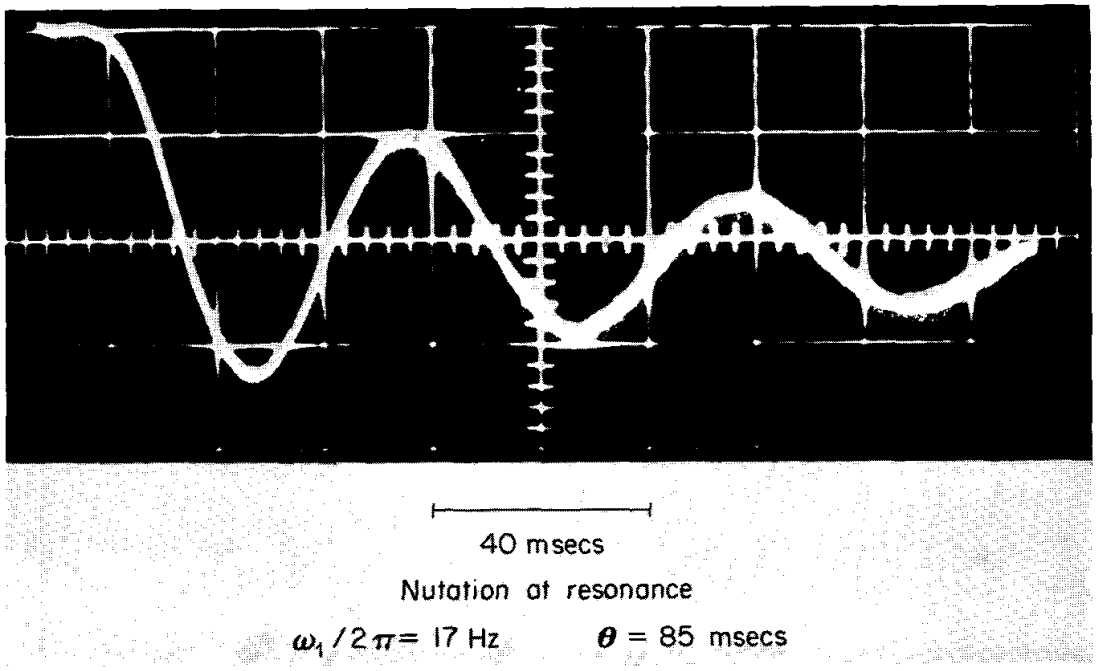
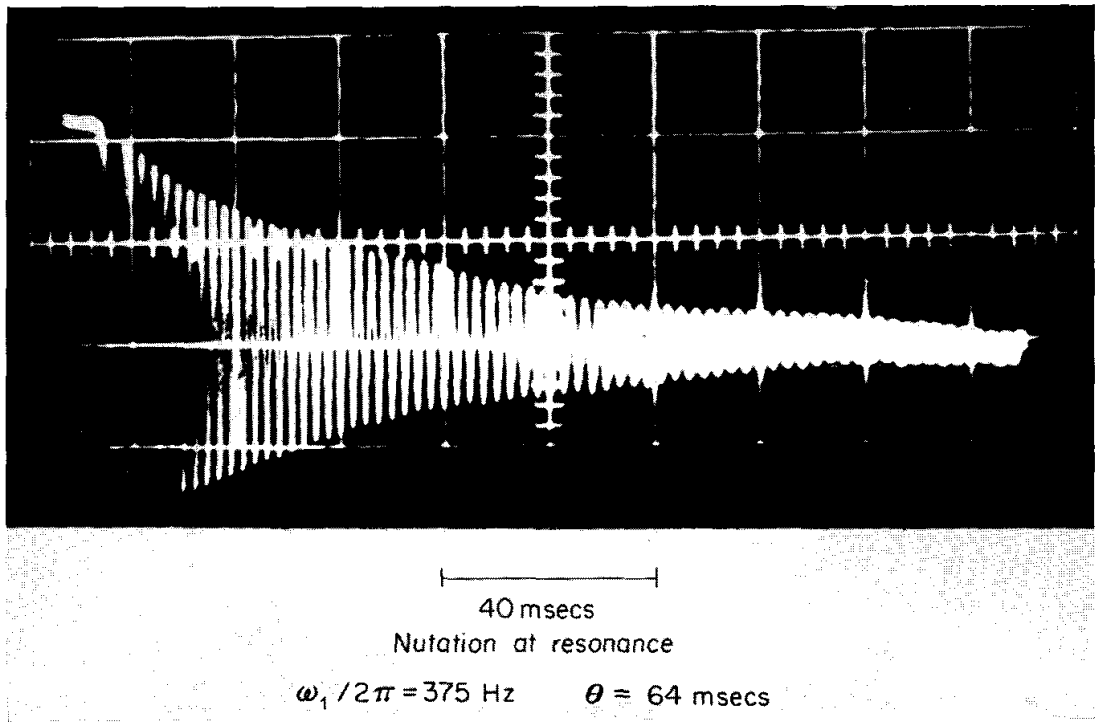


FIG. 25. Nutation at resonance of ^{3}He in the ground state.
a. Weak r.f. field.



b. Stronger r.f. field.
The time constants for relaxation can be obtained from the damping of these curves.

7.3. MONITORING LAMP

An alternative procedure is to use separate lamps for pumping and monitoring. The latter should be weak enough so that its pumping and relaxing effect is negligible. If such a lamp is used the indirect technique described above is unnecessary. The relaxation curve (T_1) can be observed directly.

7.4. NUTATION: MEASUREMENT OF τ_2

The transverse relaxation can be studied by observing the nutation as in Fig. 25. The sample is pumped to equilibrium and the r.f. field, previously adjusted to the resonance frequency, is suddenly switched on. The longitudinal magnetization nutates—i.e., precesses round the effective field which is rotating in the x–y plane (classical picture of magnetic resonance). This nutation is damped at the rate $\theta^{-1} = \frac{1}{2}(\tau_1^{-1} + \tau_2^{-1})$.

Figures 25a and b relate to ^{3}He pumped by the process of metastability exchange and monitored via the metastable state. The relaxation time constants in these examples are 64 and 85 msec. The frequency of nutation, 375 Hz in the one case and 17 Hz in the other, is, at resonance, proportional to the r.f. field strength, and measurement of this frequency is a good method for determining this field strength.

8. FREQUENCY SHIFTS

Associated with the relaxation processes we have mentioned are dispersive effects which may be manifested as frequency shifts. Thus, there is a frequency shift which arises from the irradiation of atoms by light, and one which arises through collisions between the optically pumped atoms and buffer gases. It will be appreciated that such shifts may give rise to serious errors in the precision spectroscopy to which optical pumping lends itself. On the other hand, the shifts allow a study of interaction processes.

8.1. PRESSURE SHIFTS

Figure 26 shows the shift of the zero-field resonance line of ^{133}Cs as a function of pressure of foreign gas. It will be noticed that both positive and negative shifts occur. With an appropriate mixture of gases, the shift may be eliminated.

8.2. LIGHT SHIFTS

Figure 27 shows the shift of the Zeeman resonance, $M_I = \frac{1}{2} \rightarrow -\frac{1}{2}$, in the ground state of ^{199}Hg. The central curve is the resonance obtained under ordinary optical pumping conditions (irradiation with circularly polarized ^{199}Hg resonance radiation). The curves on either side show the displacement

obtained by irradiating the vapour in addition with σ^+ or σ^- light from *a different isotope*, ^{201}Hg.

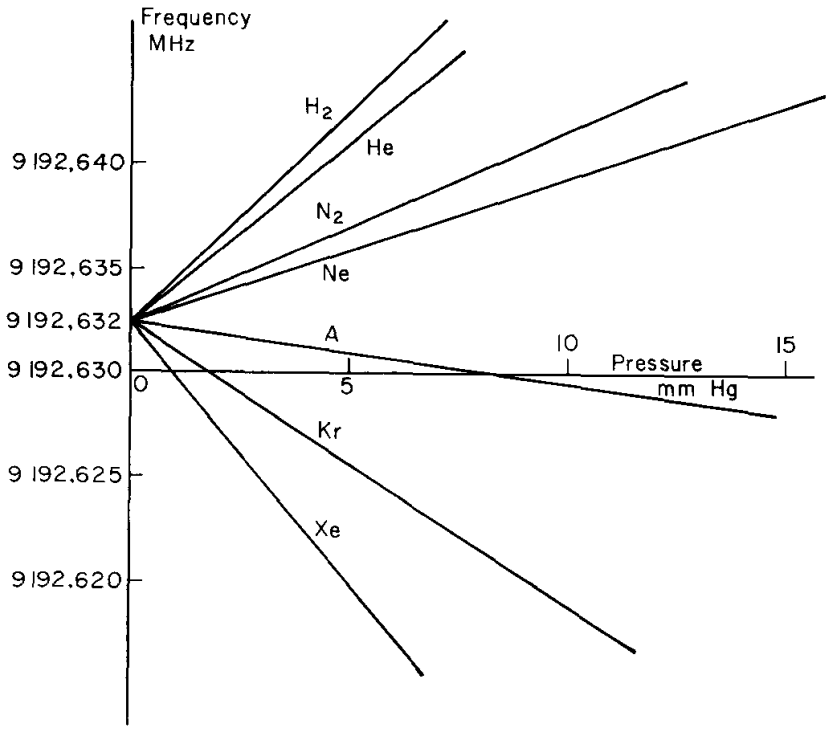


FIG. 26. Pressure shift of zero field hyperfine resonance line of ^{133}Cs for different buffer gases. (After Arditi and Carver.[83])

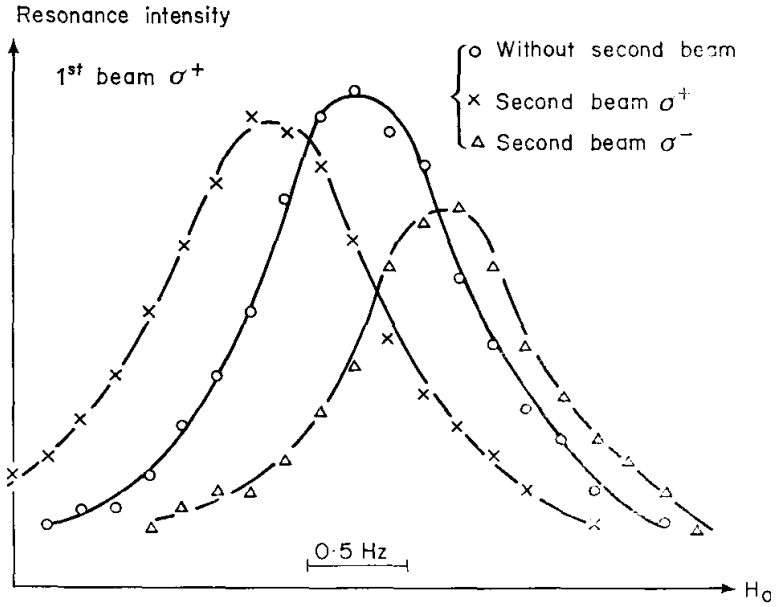


FIG. 27. Resonance curves in the ground state of ^{199}Hg demonstrating light shifts. (After Cohen-Tannoudji.[82])

One chooses light from a different isotope because the spectral components of ^{201}Hg do not coincide with the absorption region of ^{199}Hg. The significance of this is the following. The dispersion curve which is associated with

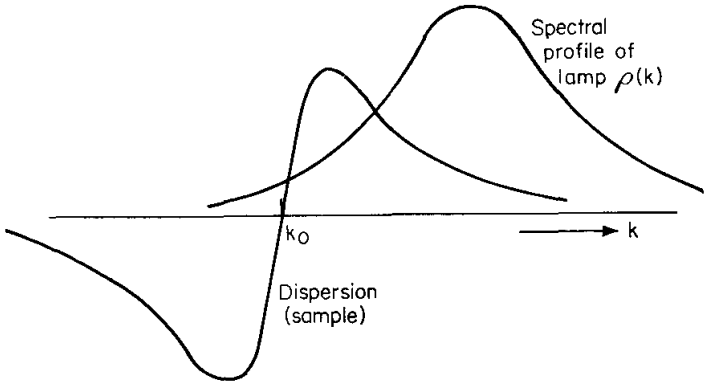


FIG. 28. The light shift is obtained by folding the spectral profile of the lamp with the dispersion curve of the sample (Section I.8.3).

absorption is antisymmetrical about the mid-point. The displacement of the energy level is given by folding the spectrum of the lamp with the dispersion curve and integrating over frequencies (Fig. 28). The result is zero if the lamp spectrum is symmetrical with respect to the absorption, but non-zero if it is asymmetrical.

The displacements seen in Fig. 27 are not the total displacements of the levels, but the *differential* displacements between the levels $M_I = \frac{1}{2}$ and $-\frac{1}{2}$.

It might appear that light shifts could be avoided by using, for irradiation of a sample, only light from the corresponding lamp, but this is not necessarily true. The spectra of pumping lamps are rarely simple lines which fall symmetrically across the absorption spectrum of the sample. Any structure in the lamp which might arise, for example, from hyperfine or isotope effects, is potentially a source of light shifts. On the other hand, the shifts themselves are small, and of significance only in precision measurements.

8.3. SEMI-CLASSICAL INTERPRETATION OF LIGHT SHIFTS AND RELAXATION

A detailed account of the optical pumping cycle has been given by Barrat and Cohen-Tannoudji.[15] But the following semiclassical interpretation of light shifts and relaxation, due to Pancharatnam[,16] is helpful.

The energy acquired by an atom of polarizability α in an electric field E is $-\frac{1}{2}\alpha E^2$. This holds also for an oscillatory field. For the complex oscillatory field $E_k \exp -ikt$ we have $\alpha_k = (e^2/2mk_0)f/(k_0 - k - \frac{1}{2}i\Gamma)$, where k_0 is a resonance frequency, Γ the corresponding damping constant, and f the oscillator strength.

The (complex) energy of interaction with radiation of energy density $\rho(k)$ is

$$\Delta W = -\sum_k \alpha_k |E_k|^2 = -2\pi \int_0^\infty \alpha_k \rho(k)\, \mathrm{d}k. \tag{9}$$

If we give this classical expression a quantum interpretation, the real part of $\Delta W/\hbar$ gives a level shift δ and the imaginary part, the damping due to the incident field. (This is the damping constant for the amplitude: the damping constant for the energy, Γ', is twice this value). Γ' should be distinguished from Γ, which is the damping constant for spontaneous emission from the excited state.

For the simple case of a transition between states $|a\rangle$ and $|b\rangle$, we have

$$f_{ba} = (2m/\hbar e^2) k_0 |P_{ba}|^2 \tag{10}$$

and

$$\alpha_{ba} = |P_{ba}|^2/\hbar(k_0 - k - \tfrac{1}{2}i\Gamma), \tag{11}$$

where P_{ba} is the matrix element of the electric dipole operator. Thus

$$\delta = \frac{-2\pi|P_{ba}|^2}{\hbar^2} \int_0^\infty \rho(k) \frac{(k_0 - k)}{(k_0 - k)^2 + (\frac{1}{2}\Gamma)^2}\, \mathrm{d}k \tag{12}$$

and

$$\Gamma' = \frac{4\pi|P_{ba}|^2}{\hbar^2} \int_0^\infty \rho(k) \frac{\frac{1}{2}\Gamma}{(k_0 - k)^2 + (\frac{1}{2}\Gamma)^2}\, \mathrm{d}k, \tag{13}$$

which are essentially the formulae derived by Barrat and Cohen-Tannoudji, illustrated in Fig. 28.

9. Level-Crossing in Ground States

An essential ingredient of the level-crossing effects in excited states (Section 4) was the preparation of atoms in superposition states. This is the condition we must look for if we wish to study level-crossing effects in ground states.

It can be achieved in the following way. Suppose we set up an optical pumping experiment to produce a polarized system. This can be done by illuminating a sample with circularly polarized light in zero field (Fig. 29).

If the system is space-quantized with reference to the direction of the light (Ox), there exists a population difference between the states, but there is no correlation between them. However, if we space-quantize with respect to another direction (Oz), each of the new states is to be expressed as a superposition of the former states, so that optical pumping along Ox has produced a superposition of states defined with reference to Oz.

If now a magnetic field is applied along Oz, the states defined with respect to Oz are energy eigenstates. They are degenerate in zero field, but the field removes the degeneracy. This is precisely the level-crossing situation we met in Section 4.

It remains to monitor the effect of removing the degeneracy. This is done by measuring the absorption of the pumping beam in the system. (It may also be done by using a separate, weak beam in the Oy direction, or by measuring the fluorescent light). One can obtain curves similar to the level-crossing curves of Fig. 14, for example, but of width corresponding to the relaxation time in the ground state.

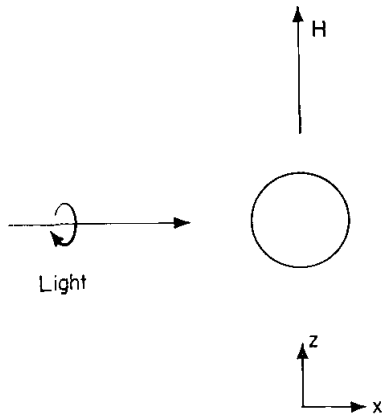


FIG. 29. Geometrical arrangement for studying the Hanle effect in absorption.

The situation we have just analysed in terms of superposition states and interference effects is sometimes described in terms of the transverse magnetisation of the system. The pumping beam creates a magnetisation along Ox. If a field H exists, the magnetisation precesses around Oz. Since different atoms are brought into the field at different times, the net magnetisation is spread out like a fan in the x–y plane. Between the extreme situations—(a) $H = 0$; magnetisation along Ox, and (b) $H \gg (\gamma\tau)^{-1}$; magnetism uniformly spread in the x–y plane—is the transition region which is precisely the region where the degenerate Zeeman levels are becoming resolved—the level-crossing region.

10. Description of Optical Pumping in Terms of Irreducible Spherical Tensor Operators

It should be remarked, in conclusion, that a description of the interaction of atoms with light in terms of magnetisation, though often extremely illuminating, is not capable of representing all the phenomena. It can represent only

those phenomena which can be analysed in terms of spherical tensors of multipole order 1. Optical interactions by electric dipole transitions can also monitor atomic properties described by tensors of multipole order 2, for which the appropriate model is a precessing and pulsating ellipsoid. Such a model has been described by Pancharatnam. The limitation to order 2 stems from the selection rule $\Delta M = \pm 1$ for electric dipole transitions, and may be understood with reference to Fig. 13. Tensors of higher multipole can, in principle, be monitored by transitions of higher multipole or by dipole transitions with more intermediate steps.

It is becoming increasingly common to analyse optical pumping experiments by means of irreducible spherical tensor operators, but no account of this work will be given in this chapter.[17, 18, 19]

II. EXPERIMENTAL DETAILS AND POINTS OF TECHNIQUE

We describe here further experimental details and points of technique.

1. Experimental Details

1.1. The sample

Two methods are in common use for containg the sample, sealed-off cells and crude atomic beams.

a. Sealed-off cells

Pyrex or silica cells are commonly used, of linear dimensions between 1 and 5 cm. Spherical or cuboid shapes are used. Sometimes the cells are extended in the form of a horn for a light trap. Good optical quality is not essential.

The vapour under study is normally at a pressure of the order of 10^{-5} or 10^{-6} torr.

Details of the application of wall coatings have been given by Bouchiat and Brossel.[20]

The vapour pressure of the sample is commonly controlled by the use of a side arm and thermostat. Special methods have been used for reactive materials. (See, for example, Minguzzi *et al.*[21])

When foreign gases are used in optical pumping experiments a wide range of pressures varying from, say, 10^{-2} torr up to pressures of the order of 1 atmosphere may be used. (Optical pumping of rubidium in argon buffer gas at pressures up to 30 atmospheres has recently been reported.[22] It is important to take account of the broadening and displacement of the absorption lines of the sample when high pressures of foreign gas are used.)

b. Atomic beams

The sample is used in the form of a beam when there is reason to fear contamination in sealed-off cells, or when it is desired to study materials which cannot conveniently be maintained as a vapour in quartz vessels. The object of the beam is not to secure collimation. Beams have been used in all-glass systems,[2] or in metal systems with windows.[23]

1.2. LIGHT SOURCES

The main requirements are that the light sources should be stable and produce a high flux in the spectral absorption region of the sample. In practically all cases this has been achieved by using a lamp whose output is a line rather than a continuous spectrum. The energy density per unit spectral range is generally insufficient in sources with a continuous spectrum. Notwithstanding this, optical pumping with white light has been performed under circumstances when the absorption line was strongly pressure-broadened,[22] and resonance fluorescence from metastable states of neon using sunlight is described by Wood in his book on Physical Optics.[24] While certain accidental coincidences of spectral lines sometimes allow the optical pumping of one element by a lamp made from another, it is most common to use in the lamp the same element that is to be studied in the sample. Use is frequently made of separated isotopes to secure differential pumping of hyperfine or Zeeman components. Very little use has been made of lasers in this field.

Insofar as the sample needs to be irradiated by light which can be absorbed by atoms in their ground or in metastable states, it follows that the spectral lines emitted by lamps of the same element are very commonly self-reversed. That is to say, the outer layers of vapour in the lamp may absorb from the broadened spectral line emitted from the core of the lamp, just those regions in the middle of the spectral profile which are needed in the sample. Much effort has gone into the production of lamps free of self-reversal though it should be appreciated that a high degree of self-reversal can be tolerated, provided there is still sufficient energy at the centre of the spectral line. Thus, many forms of commercial sodium lamp show a high degree of self-reversal in their spectrum but have nevertheless often been used for optical pumping.

The disadvantage in irradiating the sample with a large flux of light outside its absorption region (as is the case when a strongly self-reversed lamp is used) is that the stray light scattered from the walls of the resonance vessel is unnecessarily increased. This is objectionable since the main source of noise in optical pumping experiments is normally photoelectric shot noise.

The different types of lamp which have been successfully used in optical pumping experiments include hollow cathode lamps, (internal electrodes) and sealed off electrodeless lamps excited by radio-frequency or microwave oscillators. A discussion of light sources in general has been given by Budick,

Novick and Lurio.[25] Bell *et al.*,[26] describe the production of small but effective alkali metal lamps. Efficient alkali metal lamps of larger surface area have been described by Atkinson, *et al.*,[27] Burling and Czajkowski[28] and by Berdowski *et al.*[29] The lamp described in the latter paper is suitable for use in high magnetic fields.

Hollow cathode lamps of advanced design, carrying high current and allowing the application of high magnetic fields, are being used at Heidelberg by Otten and zu Putlitz.[30]

When sealed-off lamps are prepared for excitation by microwave sources the cells are normally flattened in order to reduce self-reversal. A typical capsule much used by the group in Paris is about 2 cm in diameter and 1 mm thick. Such capsules are placed near an antenna from a microwave oscillator which may typically be a magnetron delivering 100 watts at 2·5 GHz, (such units are

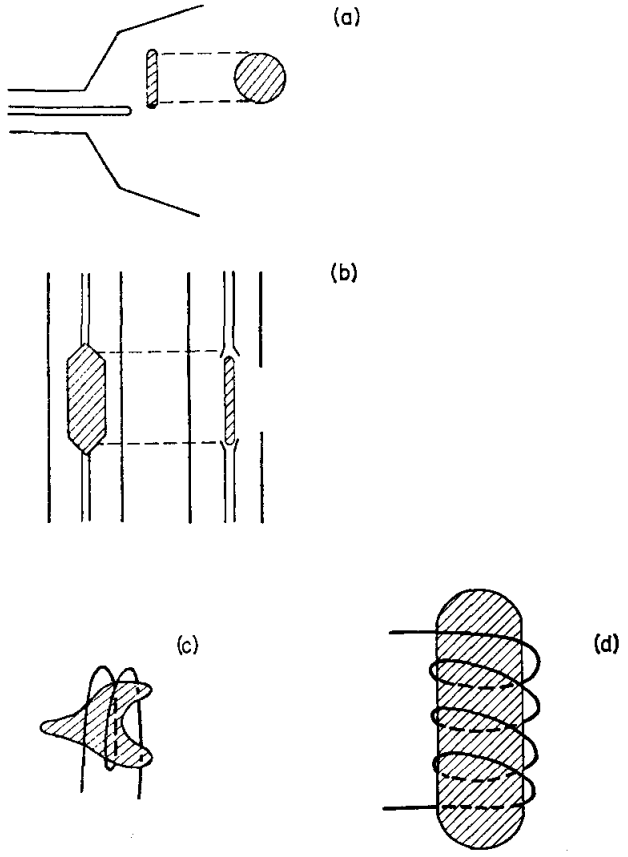


FIG. 30. Some typical light sources: sealed-off cells.

a. Disc excited by antenna from microwave oscillator.
b. Flattened cylinder forming part of co-axial line.
c. Cylinder with re-entrant window in coil of oscillator at about 100 MHz.
d. Cylinder in coil of oscillator at about 10 MHz.

produced for medical purposes). Alternatively, if the capsules are prepared in the form of a squashed cylinder a few centimeters long and a millimeter thick they may be incorporated as part of the central conductor in a co-axial line driven by a similar oscillator. Capsules used for these purposes will contain a noble gas at a pressure of about 1 torr together with the material whose spectrum is to be excited. It is sometimes necessary to provide a thermostat so that the pressure of the vapour may be controlled independently of the current in the lamp.

A microwave-excited lamp suitable for exciting calcium and magnesium ions has been described by Smith and Gallagher.[31] (See Fig. 31).

1.3. MAGNETIC FIELDS

It is rarely necessary in optical pumping experiments to use high magnetic fields; the requirements are generally to produce relatively small magnetic fields of high homogeneity. The amount of non-homogeneity which can be tolerated depends on the line width of the resonance being explored. For zero field level-crossing experiments in excited states, for example, where typical line widths might be of the order of a few gauss, stray fields in the laboratory are often a significant perturbation. For optical pumping experiments in ground states it is essential to compensate stray fields. This is commonly done

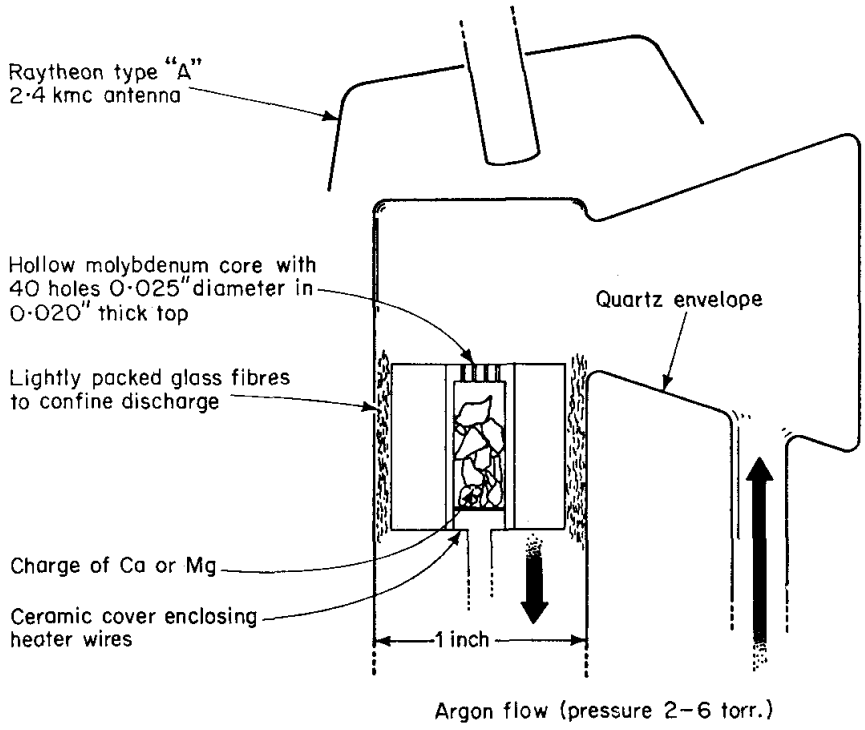


FIG. 31. Flow lamp excited by microwave antenna. (After Smith and Gallagher.[31])

with orthogonal pairs of Helmholtz coils of diameter of the order of 30 cm. Difficulties in obtaining circular formers of this size may be avoided by winding rectangular coils with formers made of channelled material.

The design of coils and solenoids to produce uniform magnetic fields has been discussed by Garrett[32] and Franzen.[33]

It is possible without too much difficulty to reduce stray static magnetic fields to a few milligauss by the use of large Helmholtz coils. Oscillatory magnetic fields (at the frequency of the A–C mains) are commonly present in the laboratory with amplitude about 2 mG. Attempts to compensate such fields by servo-mechanisms have been rather unsuccessful. More commonly these stray fields have been reduced by shielding.

In a recent experiment Dupont-Roc *et al.*[34] studied resonances of width about 1 μG for which the sample was enclosed within four concentric cylinders of Mu-metal.

1.4. DETECTORS

Photocells and photomultipliers are commonly used together with electric amplification and recording, although in some of the early work sensitive galvanometers were used very successfully. Many investigations have been successfully performed with a single photodetector. This is possible only when one can rely on the stability of the lamp.

Compensation for drifts in the lamp can often be effected by taking fluorescent light into two photodetectors in such positions that the two signals are in opposite senses. By the use of an optical bridge or by some other arrangement, the signal can be doubled and the effects of drifts eliminated to a first approximation.

In simple cases level-crossing and double resonance signals can be a substantial fraction of the total fluorescent light. In such cases the signal can be displayed on an oscilloscope with wide-band amplification. More commonly narrow band methods of detection are used to rescue the signal from noise: a typical system would use phase sensitive detection with time constants of a few seconds, but in the more complicated cases which are currently being studied the signal is a very small fraction of the fluorescent light and signal averaging methods are increasingly being used.

A novel method of detection, applicable to short-lived radioactive elements, is described in a later section (II 6.4).

1.5. FILTERS AND POLARIZERS

It has been explained that optical pumping is sometimes more advantageously carried out by filtering some of the components of the light from the lamp. Interference filters are commonly used to separate the alkali doublets, for

example. Lyot (polarization) filters are also used for this purpose. Hyperfine components can sometimes be suppressed by introducing into the pumping beam a cell of a different isotope of the element concerned, to act as a differential absorber.

Sheet polarizers (linear and circular) are often adequate but there are occasions when more complete polarization is required and in such circumstances crystal polarizers are used. While one-beam polarizers (e.g. Glan prisms) are attractive, greater aperture can be achieved with two-beam prisms (e.g. Rochon). The unwanted beam is focussed and rejected by a stop.

2. Spectrum of the Lamp

Our analysis, in Section I, of double resonance and level-crossing experiments was based on the assumption that the spectrum of the lamp was uniform across the whole of the absorption region of the sample. There are occasions when this approximation is no longer valid, in particular when the absorption spectrum is being shifted by the application of magnetic fields. In such cases the profile of the resonances may be distorted. This is particularly serious when line profiles are being studied with a view to measuring lifetimes. The resonance line profiles may then be calculated by an analysis based on resolution of the spectrum of the lamp into its Fourier components. The signal due to monochromatic components interacting with stationary atoms is then to be integrated over the spectrum of the lamp and the Doppler absorption profile of the sample.[35, 36]

3. Excitation by Electrons and other Particles

There are sometimes advantages to be gained by exciting atoms with electrons or other particles rather than by light. So long as some departure from the statistical population of excited states can be achieved by such methods of excitation, radio-frequency resonance and level-crossing phenomena can be monitored by changes in the fluorescent light.

3.1. electrons

Electron excitation has been developed particularly by Pebay-Peyroula and his colleagues.[37] An account of recent work in this field is given in the Proceedings of a Conference held recently in Grenoble.[38]

A greater variety of excited states can be reached by electron excitation as compared with optical excitation, but there are two major difficulties in the use of electrons. One is that the fluorescence being detected is often affected

by decay from higher levels in cascade. The other is that the electron trajectories are perturbed by the applied magnetic fields which by the nature of the experiments are required to be varied. Very pronounced resonance effects occur when the cyclotron frequencies of the electrons in the static field approach the frequency of the oscillating field. In order to avoid these effects it is arranged, where possible, that the electrons should travel parallel to the static field and that the path length should be such that off-axis electrons are unable to perform a complete cycle of cyclotron motion in the course of their flight.

Level-crossing experiments require that the electron beam be transverse to the magnetic field (this is a symmetry requirement corresponding to the optical arrangement in which the incident light is polarized at right angles to the magnetic field). Here the deflection of the beam by the magnetic field is particularly serious. The change in deflection as the field is varied gives rise to distortion of the level-crossing line shape.

While most experiments with electron excitation have been performed with a heated filament as a source of electrons, some recent work has been performed in sealed-off vessels where the electrons have been generated in a gas discharge at low pressure. The vessel is placed between condenser plates at the end of Lecher wires. The motion of the electrons under these circumstances is dominantly an oscillation, perpendicular to the plates.[39]

3.2. HEAVY PARTICLES

The use of heavy particles rather than electrons reduces by many orders of magnitude the difficulties connected with the deviation of electrons by the applied magnetic fields. Level-crossing experiments in helium excited by H_2^+ have been successfully carried out by Kaul.[40] A quantitative study of the polarization of fluorescent light from helium excited by protons has been made by Krause and Soltysik.[41]

Selective excitation can also be achieved in beams of fast particles which have traversed a thin foil.

4. COHERENCE NARROWING, PRESSURE BROADENING AND DEPOLARIZATION

Our discussions of double resonance and level-crossing experiments have hitherto supposed that the signals correspond to the single scattering of light by atoms. If the vapour pressure is low enough the experimental results do, in fact, correspond to single scattering, but as the vapour pressure is increased the light in the resonance vessel may undergo multiple scattering. The first consequence of this is that the light becomes depolarized with consequent loss of signal. But strong signals may nevertheless still be obtained and it is found that, contrary to expectations, the resonance curves and level-crossing curves

become narrower with increase of vapour pressure. As the pressure is increased still further the curves begin to broaden owing to the reduction in lifetime of the excited atoms through collisions.

The narrowing was discovered by the group in Paris and interpreted by them as a consequence of the coherent diffusion of light through the vapour. The phenomenon is called "coherence narrowing". A detailed experimental and theoretical study was made by Barrat.[42] It has since been discovered that the coherence narrowing is accompanied by a shift in the magnetic resonance frequencies.[43, 43a, 44]

It is necessary to be aware of the phenomenon of coherence narrowing when experiments are undertaken to determine the lifetime of excited states from level-crossing curves. Such curves are taken at a range of vapour pressures and the width, plotted as a function of atomic density, extrapolated to zero density. The broadening at higher vapour pressures allows a study of interatomic interactions. (See, for example, Happer and Saloman[17]; Omont and Meunier.[18])

5. Comparison of Double Resonance and Level-Crossing Experiments: Modulation Techniques

While double resonance experiments have been extensively applied in studies of excited atoms, they suffer from serious disadvantages which arise from the need to use a strong oscillating field on the sample.

(i) It is not always a simple matter to achieve the necessary amplitude of magnetic field at the high frequencies required.

(ii) The oscillating field perturbs the resonances in two ways; first by broadening them and secondly by shifting their positions. (See Section IV).

(iii) The strong oscillating fields sometimes set up gas discharges in the resonance vessels and the light from such discharges, though weak, is sometimes sufficient to swamp the fluorescent light.

For these reasons level-crossing experiments are to be preferred. The sole external perturbation on the excited atoms is then simply the beam of light used for excitation. Level-crossing experiments can give values for lifetimes of excited states accurate to 1 or 2% in favourable cases. Measurements to this precision demand a careful study of the line profile, which is of the pure Lorentzian shape only for certain well-defined geometrical conditions. The use of finite light cones may introduce some mixture of the dispersion shape—a point which must be carefully studied.

Measurements of the position of level-crossing curves yield values for the hyperfine interaction constant which are fundamentally limited only by the natural width of energy levels. However, calculation of these interaction constants from the measured magnetic fields at which the level-crossing signals are found depends on a knowledge of the g_J-factor of the states, and if this is not known from optical measurements it must be determined by a double resonance experiment. Indeed, the determination of g_J-factors is now the most important application of the double resonance method.

An alternative to the double resonance method presents itself in the use of modulated light. If a level-crossing experiment is performed with modulated light the level-crossing signals are displaced to the position where the modulation frequency matches the interval between the levels.[45] From the measured fields and frequencies the g-value can be deduced. This is a type of double resonance experiment which requires no oscillatory field to be applied to the sample. The curves obtained are identical with level-crossing curves—that is to say they suffer neither from radio-frequency broadening nor from displacement of the resonances. The technical difficulties are now transferred to the means of modulating the light. Because of these difficulties the method has not been widely applied. It has, however, been used to verify the physical reality of the frequency diagram—a representation of combinations of the nutational and precessional frequencies in the motion of atoms under oscillating fields (See Section III). The frequency diagram for the excited $^3P_1$ state of mercury has been explored in this way.[46,47]

6. Some Typical Results and some Recent Applications of Optical Pumping, Double Resonance and Level-Crossing Techniques

No attempt will be made here to give a complete account of work in this field. Some recent compilations have been made by zu Putlitz,[48] by Budick,[49] and Major.[50] The experiments described in the following sections have been chosen to illustrate a variety of applications.

6.1. Hyperfine structure of hydrogen

As an example of the application of optical pumping to measure quantities of fundamental importance in atomic physics, the series of experiments by Pipkin and his colleagues[51] on the hyperfine structure of hydrogen, deuterium and tritium may be cited. The measurements of the hyperfine intervals were made to a precision of a few hertz, the intervals themselves being of the order of 10^9 Hz. Knowledge of these intervals is of the greatest importance in verifying theories of atomic structure and radiation interactions and in determining the value of the fine structure constant.

6.2. PRECISION MEASUREMENT OF ATOMIC g-FACTORS

As an example of more recent precision measurements in optical pumping we refer to the work of White *et al.*[52] on the g-factor ratios for free rubidium atoms. Precision measurements of such quantities were formerly made by the methods of atomic beam resonance spectroscopy. Optical pumping methods offer an alternative technique. In the work of White *et al.* the ratios g_I/g_J were measured for ^{85}Rb and ^{87}Rb to a few parts per million and the ratios $g_J(^{87}\text{Rb})/g_J(^{85}\text{Rb})$ to a precision of a few parts in 10^9. These results were obtained from evacuated wall-coated cells. Distortion of the Lorentzian line shape was found for cells filled with buffer gases. The results were corrected for shifts arising from the pumping light.

6.3. RELAXATION STUDIES BY OPTICAL PUMPING

An extended series of experiments on the relaxation of optically pumped rubidium has been carried out by Mme Bouchiat. A recent report[20] gives an account of the relaxation of rubidium on paraffin coated walls. The results are compatible with other measurements on the interaction of rubidium atoms with buffer gases.

6.4. DETECTION OF OPTICAL PUMPING BY RADIOACTIVE EMISSION

A variation of the optical pumping method has recently been used to find the nuclear spins and moments of certain short-lived radioactive isotopes.[53] It is applicable to isotopes which can be produced by the bombardment of a gas by particles from an accelerator and whose lifetime is short enough for them to decay before they have had time to diffuse through the gas to the walls of the containing vessel.

The isotopes produced in this way are polarized by circularly polarized light, either directly or through spin-exchange. Their polarization is monitored by measuring the asymmetry in the β–emission. The polarization is destroyed by a radio-frequency field at resonance with the Zeeman or hyperfine intervals and the signal is detected by a change in the counting rate.

6.5. LEVEL-CROSSING EXPERIMENTS ON HYDROGEN

The fine structure of excited states of atomic hydrogen is no less important than the hyperfine structure in atomic physics. By the application of magnetic fields of a few thousand gauss the spin-orbit coupling is broken down and Zeeman levels of $2P_{3/2}$ intersect those of $2P_{1/2}$. A measurement of the crossing point allows a determination of the $P_{1/2}-P_{3/2}$ interval and of the fine structure constant. A level-crossing experiment to determine these quantities was recently reported by Metcalf *et al.*[54] Similar studies have also been undertaken by Fontana and colleagues.[55,56] The sharpness of the resonances is less than

in the measurement of hyperfine structure because it is limited by the short lifetime of the excited P-states. The fine structure constant is determined to a precision of about one part in 10^5.

6.6. LEVEL-CROSSING EXPERIMENT ON MANGANESE

An example of an experiment in which a weak signal from a complicated structure was analysed to determine the hyperfine structure is the level-crossing experiment of Handrich *et al.*,[57] on the MnI spectrum. The modulation necessary for phase sensitive detection was achieved by rotating a polarizer in the fluorescent beam. The signal was retrieved from noise by the use of an Enhancetron.

6.7. HYPERFINE STRUCTURE OF EXCITED ATOMS

As an example of a recent application of double resonance methods to the measurement of a hyperfine structure, we cite the work of Feiertag and zu Putlitz.[58] This study of the $7^2P_{1/2}$ term of ^{85}Rb was of value in assessing the contribution of core polarization to the magnetic hyperfine structure.

6.8. MEASUREMENT OF g_J-FACTORS OF EXCITED STATES

The structure of levels of excited atoms is of interest not only from the point of view of hyperfine structure but also because measurements of g_J-values allow one to assess the degree of breakdown of L–S coupling, for example, and to determine whether or not configuration interaction needs to be taken into account. In this context the double resonance method was used by Ma *et al.*,[59] to measure the g-factors of low-lying $^3P_1$ states of strontium and barium. The g-factors were measured to a precision of a few parts in 10^5.

7. APPLICATION OF OPTICAL METHODS TO MOLECULES AND IONS IN SOLIDS

We conclude this section with a brief reference to work on systems other than atoms.

7.1. MOLECULES

A detailed analysis of the possibilities for molecular level-crossing spectroscopy has been given by Zare.[60] Crosley and Zare[61] applied this method to nitric oxide. The width of the zero field level-crossing curve for the state investigated was about 0·4 G, but this cannot be interpreted as a lifetime until the g-value is known. A double resonance experiment on the same state allowed the determination of the g-factor and hence the lifetime.[62]

In the case of molecules far more than in atoms, accidental coincidences of wavelength allow the excitation of the molecule by line radiation from a lamp of a different substance.

7.2. PARAMAGNETIC IONS IN SOLIDS

As an example of the application to ions in solids, we refer to a recent paper by Imbusch *et al.*[63] on excited states of V^{2+} and Mn^{4+} in Al_2O_3. This work allowed the measurement of hyperfine structure and spin-lattice relaxation of excited ionic states. References to earlier work in this field are given in the paper. More recently it has been shown by Chase[64] that double resonance applied to excited states of ions leads to modulation in the fluorescent light, just as in the case of free atoms.

III RADIO-FREQUENCY INTERACTIONS

(a) COHERENCE AND OPTICAL MONITORING

1. Distinction Between Optical Fields and Radio-Frequency Fields

There is an important distinction to be drawn between the interaction of atoms with fields of extended spectral range (broad-band) on the one hand and of sharp frequency on the other. In the context of optical pumping experiments the interactions with light (including spontaneous emission) come under the former heading and with radio-frequency fields under the latter. For broad-band interactions the transition probabilities are independent of time and the absorption and emission processes can be represented by exponentials. Such processes are called "rate processes". In the Einstein treatment of the interaction of radiation with atoms, absorption, stimulated emission and spontaneous emission are treated as rate processes. For monochromatic fields on the other hand the transition probabilities are time-dependent, leading to a sinusoidal oscillation of population between two states connected by the perturbation. This sinusoidal oscillation is formally called nutation, but in the context of magnetic resonance one commonly speaks of spin-flipping. It is important to recognise the existence of two distinct frequencies in the context of magnetic resonance: the driving frequency whose phase is determined by the phase of the laboratory oscillator, and the nutational or spin-flip frequency for which the phase depends on the moment at which the system is introduced into the field and which in steady state experiments is a random variable.

It is, of course, possible to obtain the result for fields of extended spectral range by integrating over the results for monochromatic fields. It is not difficult to show analytically—and, indeed, the conclusion is very plausible—that the response of the system is exponential rather than sinusoidal if the amplitude of the initial state (in the interaction representation) does not change appreciably in the coherence time (reciprocal of the spectral range) of the exciting radiation. From this result one may deduce the following condition for the exponential response: the spectral range of the exciting radiation must be greater than the perturbation (measured in frequency units) applied to the final state of the transition. This perturbation may be that which arises from the exciting field itself or it may be some other purturbation such as an additional relaxing process.

2. Monochromatic Fields: Semiclassical or Quantized Field Treatment?

In the semiclassical treatment one describes the field classically and the matter quantum mechanically. The field variables are treated as ordinary algebraic quantities (c-numbers) in the analysis, but operators are used to describe dynamical variables of the atoms.

In the quantum field theory the radiation is quantized as well as the matter and the field variables are operators. The state functions are functions both of the field and of the matter.

The important difference between the two approaches lies in the treatment of the spontaneous emission. This needs no special consideration in the quantized field method, but needs supplementary assumptions in the semi-classical method. We shall state these assumptions in due course when we need to use them. All we need at the moment is to know under what circumstances the two treatments are equivalent.

They are equivalent (if properly formulated) when spontaneous emission can be ignored. This is ordinarily true in magnetic resonance or in other situations when the field is monochromatic and the mean photon-number is large compared with unity, for in this case the probability for spontaneous emission is much less than the probabilities for absorption and stimulated emission. This is an example of the normal condition under which quantum theory goes over into classical theory, namely that the contemplated change in a quantum number (in this case the photon-number) is large in comparison with the value of that number. But it is worth noticing that the quantized field and semiclassical theories are also equivalent for absorption processes treated by first order perturbation theory even when the optical fields are weak, (mean photon-number per mode much less than unity) for when one is dealing with absorption the possibility of spontaneous emission is irrelevant.

In the quantized field theory, as normally written, the basis states are energy eigenstates of the system (atoms + optical field), and the radio-frequency field is treated classically. In a recent formulation, Cohen-Tannoudji and Haroche[65] have taken (atoms + radio-frequency field) as the closed system whose energy eigenstates form the basis states. Time-dependence is treated by using superposition states.

In the semiclassical theory, time dependences, both of the optical and radio-frequency fields, appear as c-numbers, and it is very easy to follow phase factors from the fields to the atomic wave function and back to the fields.

We shall use the semi-classical treatment in this account. Detailed analyses will be restricted to studies of the radio-frequency interactions. We shall avoid difficulties connected with spontaneous emission by making use of the "pulse approximation", and we shall generally use the pulse approximation also in dealing with the absorption of light.

3. Monochromatic Fields: Semiclassical Treatment

Two cases can be solved exactly:

(i) rotating field in the plane perpendicular to a static field.

(ii) field oscillating parallel to a static field (provided that the atomic coupling is not broken).

The methods of solution are different for these two cases. We shall treat the first fairly thoroughly and the second briefly, in Section III.7. We shall be thinking of magnetic fields, though much of the work will be applicable to electric fields also.

3.1. A field rotating in the plane perpendicular to a static field

(This is the standard arrangement for magnetic resonance. The problem and its solution are well known but we wish to establish a notation and to derive some results for reference.)

$$\text{Let the field be } H\mathbf{k} + H_1 (\cos \omega_0 t\, \mathbf{i} + \sin \omega_0 t\, \mathbf{j}), \tag{14}$$

where **i**, **j**, **k** form a set of Cartesian unit vectors. Let us write for the Hamiltonian of the system $\mathscr{H}_0 + \mathscr{H}_1$, where $\mathscr{H}_0$ includes all time independent and isotropic interactions in addition to the interaction with the static field H and $\mathscr{H}_1$ describes the interaction with the transverse field H_1.

The method of solution is:

(i) make a transformation to eliminate the time dependence;

(ii) integrate the equation of motion and write in the time development;

(iii) Diagonalize the transformed Hamiltonian;

(iv) transform back to the initial frame.

This sequence of operations yields the operator which gives the development in time of the state vector or density matrix of the atomic system (time-displacement operator). The matrix elements of the operator are easily found and the transition probabilities readily obtained.

Let us study the steps (i) to (iv) in turn and then apply the method to a simple case.

(i) The operator which removes the time dependence is $\exp(i/\hbar)\, J_z\, \omega_0\, t$ where J_z is the operator representing the component of angular momentum J along the direction of the static magnetic field (z–axis). As is well known this operator transforms the Hamiltonian $\mathscr{H}_0 + \mathscr{H}_1$ to

$$\mathscr{H}' = \mathscr{H}_0 + \mathscr{H}_1' - \omega_0 J_z, \tag{15}$$

where $$\mathscr{H}_1' = \exp[(i/\hbar) J_z\, \omega_0\, t]\, \mathscr{H}_1 \exp[-(i/\hbar) J_z\, \omega_0\, t].$$

We shall apply this transformation first at the particular time $t = t_0$ at which the atoms begin to experience the field, and secondly at the later time t at which we sample the system. For a simple spin system the transformation is equivalent to changing the field given by eqn (14) to the effective field

$$\mathbf{H}_{\text{eff}} = (H - \omega_0/\gamma)\mathbf{k} + H_1\, \mathbf{i} \tag{16}$$

(γ is the gyromagnetic ratio).

(ii) The equation of motion is

$$i\hbar \frac{\partial}{\partial t} |'t\rangle = \mathscr{H}' |'t\rangle, \tag{17}$$

where $|'t\rangle$ is the state vector at time t in the transformed frame. Since $\mathscr{H}'$ is independent of time the equation integrates to

$$|'t\rangle = \exp[-(\mathrm{i}/\hbar)\mathscr{H}'(t - t_0)]|'t_0\rangle. \tag{18}$$

We can most easily apply this equation if $\mathscr{H}'$ is diagonalized. This leads us to the next step.

(iii) Diagonalization of the transformed Hamiltonian is easily accomplished for the simple spin system by choosing a representation in which the axis of quantization is along $\mathbf{H}_{\text{eff}}$, i.e., by a rotation of Oz through the angle β about the x–axis from the direction of H to the direction of $\mathbf{H}_{\text{eff}}$.

$\beta = \tan^{-1} H_1/(H - H_0)$. The operator is the rotation matrix $\mathscr{D}^{(\mathrm{J})}(0, \beta, 0)$.

In a more complicated system where the fields are strong enough to introduce some decoupling of spins we suppose that the Hamiltonian is diagonalized by an operator S.[66]

In either case let us suppose that the eigenvalues of the diagonalized Hamiltonian are λ_i, that is to say the λ_i are the eigenvalues of $S\mathscr{H}'S^{-1}$ or of $\mathscr{D}(\beta)\mathscr{H}'\mathscr{D}(-\beta)$.

We now make use of the relation

$$\exp -(i/\hbar)\mathscr{H}'(t-t_0) = S^{-1}\{\exp-(i/\hbar)S\mathscr{H}'S^{-1}(t-t_0)\}S, \tag{19}$$

and notice that the time development of $\mathscr{H}'$ has been replaced by the time development of $S\mathscr{H}'S^{-1}$ whose eigenvalues will appear as the frequencies in the periodic motion. These eigenvalues are the nutational frequencies.

(iv) The final transformation is to apply $\exp[-iJ_z\omega_0 t]$ which takes us back to the laboratory frame at time t.

By this sequence of operations we have found the form of the time-displacement operator $U(t, t_0)$ defined by

$$|t\rangle = U(t,t_0)|t_0\rangle. \tag{20}$$

We have

$$U(t, t_0) = \{\exp-(i/\hbar)J_z\,\omega_0\,t\}\,S^{-1}\{\exp-(i/\hbar)S\mathscr{H}'S^{-1}(t-t_0)\} \times S\{\exp(i/\hbar)J_z\,\omega_0\,t_0\}. \tag{21}$$

a. Example

A particle with spin-$\frac{1}{2}$. In the field H alone, let the eigenstates be $|+\rangle$, $|-\rangle$. The energy eigenvalues are $\pm\frac{1}{2}\hbar\omega$, with $\omega = \gamma H$. Probability amplitudes are a_+, a_-. Suppose we are given $a_+ = 1, a_- = 0$ at time t_0. The problem is to find the values of a_+ and a_- at later time t.

The state vector at time t_0 is $\binom{1}{0}$. Using (20) and (21) we can immediately write down

$$\begin{pmatrix} a_+(t) \\ a_-(t) \end{pmatrix} = \begin{pmatrix} \exp-\frac{1}{2}i\omega_0 t & 0 \\ 0 & \exp+\frac{1}{2}i\omega_0 t \end{pmatrix}\begin{pmatrix} S^{-1}_{++} & S^{-1}_{+-} \\ S^{-1}_{-+} & S^{-1}_{--} \end{pmatrix} \times \begin{pmatrix} \exp-\frac{1}{2}ip(t-t_0) & 0 \\ 0 & \exp+\frac{1}{2}ip(t-t_0) \end{pmatrix} \times \begin{pmatrix} S_{++} & S_{+-} \\ S_{-+} & S_{--} \end{pmatrix}\begin{pmatrix} \exp+\frac{1}{2}i\omega_0\,t_0 & 0 \\ 0 & \exp-\frac{1}{2}i\omega_0\,t_0 \end{pmatrix}\begin{pmatrix} 1 \\ 0 \end{pmatrix}. \tag{22}$$

We have used the values $\pm\frac{1}{2}\hbar$ for the eigenvalues of J_z and $\lambda_\pm = \pm\frac{1}{2}\hbar p$ for the eigenvalues of $S\mathscr{H}'S^{-1}$. If for this case, we identify S with $\mathscr{D}^{(\frac{1}{2})}(\beta)$, we have

$$\begin{pmatrix} S_{++} & S_{+-} \\ S_{-+} & S_{--} \end{pmatrix} \equiv \begin{pmatrix} \cos\beta/2 & \sin\beta/2 \\ -\sin\beta/2 & \cos\beta/2 \end{pmatrix} = \begin{pmatrix} c & s \\ -s & c \end{pmatrix}. \tag{23}$$

Performing the matrix multiplication we find

$$\begin{pmatrix} a_+(t) \\ a_-(t) \end{pmatrix} = \begin{pmatrix} \exp -\frac{1}{2}i\omega_0 t[c^2 \exp -\frac{1}{2}ip(t-t_0)+s^2 \exp +\frac{1}{2}ip(t-t_0)] \\ \exp +\frac{1}{2}i\omega_0 t[sc \exp -\frac{1}{2}ip(t-t_0)-sc \exp +\frac{1}{2}ip(t-t_0)] \end{pmatrix}$$

$$\times \exp +\tfrac{1}{2}i\omega_0 t_0, \quad (24)$$

whence $$|a_+(t)|^2 = 1-\tfrac{1}{2}\sin^2\beta[-\cos p(t-t_0)], \quad (25)$$

$$|a_-(t)|^2 = \tfrac{1}{2}\sin^2\beta[1-\cos p(t-t_0)], \quad (25a)$$

and

$$a_+a_-^* = \tfrac{1}{2}\sin\beta\{c^2[1-\exp -ip(t-t_0)]-s^2[1-\exp +ip(t-t_0)]\}\exp -i\omega_0 t. \quad (25b)$$

$$\tan\beta = H_1/(H-H_0), \text{ where } H_0 = \omega_0/\gamma.$$

Equations (25) and (25a) give the populations of the states. Notice the sinusoidal variation with frequency p. Notice also that the driving frequency ω_0 does not appear in these equations.

Equation (25b) gives the correlation between the states. The driving frequency appears in this equation as well as the nutational frequency, p. Notice that p occurs with the time interval $(t-t_0)$, whereas ω_0 occurs with the time t.

b. Frequency diagram

Let us study the periodic terms which occur in equation (24). Firstly, the common factor $\exp +\frac{1}{2}i\omega_0 t_0$ is irrelevant. It represents an arbitrary phase factor common to both states. Next, we see the factors $\exp \pm\frac{1}{2}i\omega_0 t$. These are periodic functions at frequencies $m\omega_0$, where m is the space quantum number with respect to the direction of the field H. But each of these terms is multiplied also by periodic terms with frequencies $\pm\frac{1}{2}p$. The $\pm\frac{1}{2}$ in these factors is the space quantum number with reference to the direction of $\mathbf{H}_{\text{eff}}$.

These frequencies and the corresponding quantum labels can be given a convenient diagramatic representation.

Figure 32a shows the energy levels of the states $|+\rangle$, $|-\rangle$ as a function of the magnetic field H. The levels are described by the equations $\omega = \pm\frac{1}{2}\gamma H$. (It is convenient to speak of the energy at this stage although the equation we have written is for the corresponding Bohr frequency).

Figure 32b shows the energy levels seen in the rotating frame, still plotted against the laboratory field H and supposing that the transverse field H_1 has not yet been applied.

Figure 32c shows the energy levels for a finite value of H_1. These are the eigenvalues $\lambda_\pm$ of $\mathcal{D}(\beta)\mathcal{H}'\mathcal{D}(-\beta)$. They are described by the equation

$$p = \pm\tfrac{1}{2}\gamma H_{\text{eff}} = \pm\tfrac{1}{2}\gamma[(H-H_0)^2+H_1^2]^{\frac{1}{2}}. \quad (26)$$

But we must remember that the eigenstates are now described with reference to the rotated axis. In order to emphasise this the states have been labelled $\oplus$ $\ominus$ These states are superpositions of the original states as follows:

$$|\oplus\rangle = \cos\beta/2\,|+\rangle + \sin\beta/2\,|-\rangle,$$
$$|\ominus\rangle = -\sin\beta/2\,|+\rangle + \cos\beta/2\,|-\rangle. \tag{27}$$

Figure 32d shows the energy levels after the transformation back to the laboratory frame. The level labelled $\oplus$ gives rise to two levels displaced by $\pm\frac{1}{2}\omega_0$, corresponding to the eigenvalues $\pm\frac{1}{2}$ of J_z for the states $|+\rangle$, $|-\rangle$. Similarly the level labelled $\ominus$ in Fig. 32c gives two levels in Fig. 32d. In Fig.

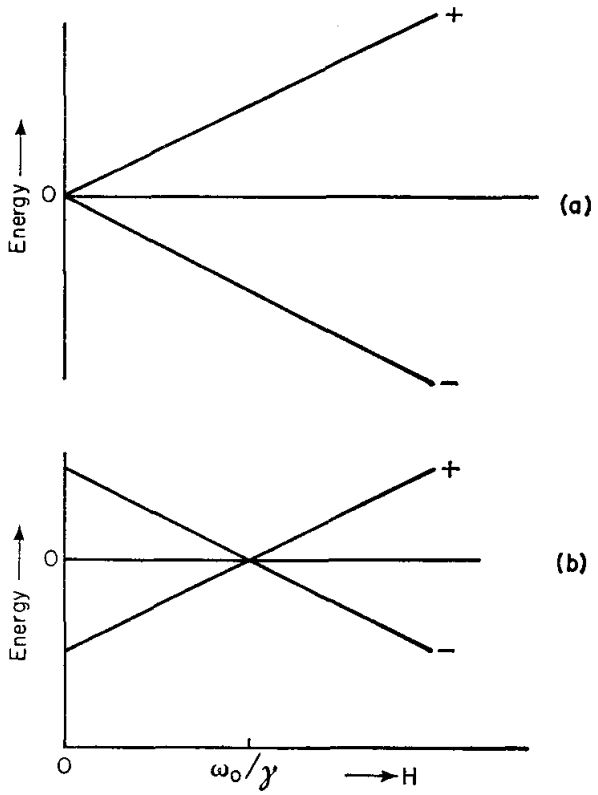


FIG. 32a. Energy of spin-$\frac{1}{2}$ system as function of magnetic field.

b. Energy of spin-$\frac{1}{2}$ system in rotating frame (angular frequency ω_0: rotation parallel to H).

32d the interval between the pairs of levels labelled $+, \oplus$ and $-, \oplus$ or $+, \ominus$ and $-, \ominus$ is ω_0, whereas the interval between a pair labelled $+, \oplus$ and $+, \ominus$ or $-, \oplus$ and $-, \ominus$ is p.

Diagrams such as Fig. 32d are generalizations of ordinary term diagrams. An ordinary term diagram is a representation of the energies of different modes of a system where the Hamiltonian is time independent. The frequency

diagram shows the side bands which appear when a periodic perturbation is applied. The levels have the same physical reality as do the energy levels in an ordinary term diagram. They may be explored, as we shall see, by suitable monitoring of the system.

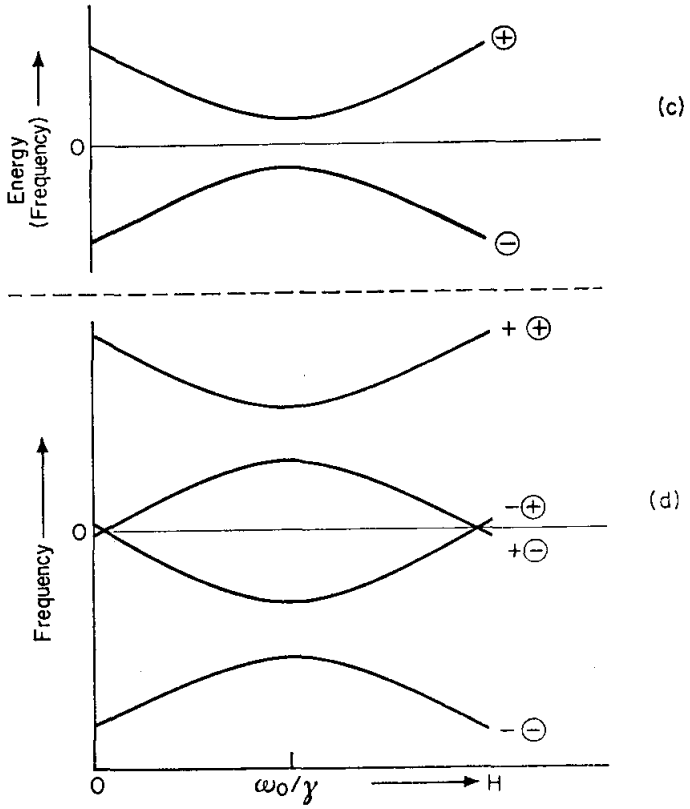


FIG. 32c. Energy in rotating frame: addition of transverse field H_1.
d. Frequency diagram in laboratory frame.

In general the nutational frequencies do not appear in steady state experiments but may be found in pulse or modulation experiments.

4. Application to Double Resonance Experiments

In such experiments the considerations of the previous section apply to the excited states. Atoms are excited by broad band irradiation, experience the interaction with the magnetic fields, and decay by spontaneous emission of light. In a steady state experiment the irradiation with light is continuous and one needs to justify the separation of the interaction into the three parts: excitation, radio-frequency interaction, and spontaneous emission. The justification lies in a comparison of the coherence times of the optical and radio-frequency perturbations with the mean lifetime of the excited atoms. For broad band irradiation the coherence time of the light is typically 10^{-10} sec,

whereas the mean lifetime is typically 10^{-8} sec. Thus, phase continuity under irradiation is lost in a time short compared with the mean lifetime and it is a good approximation to consider the excitation as a random sequence of pulse processes. The coherence time of the radio-frequency interaction, on the other hand, is, for a continuous wave oscillator, the period for which the oscillator is switched on, so that continuity of phase of the radio-frequency perturbation is something that cannot be ignored. As for spontaneous emission, it is convenient to think of this as the result of a perturbation stimulated by random fluctuations having the spectrum of black body radiation at the absolute zero. For such a spectrum the coherence time needs careful definition, but it suffices here to think of the coherence time as the reciprocal of the frequency which is being stimulated. The time we have to consider therefore is of order 10^{-15} sec which allows spontaneous emission also to be treated as a pulse process.

Our model is therefore as follows:

(i) an atom is excited at time t_0;

(ii) its wave function evolves in the excited state under the influence of the magnetic fields. It is nearly always true in double resonance experiments that the wavelength of the radio-frequency field is large compared with the size of the experimental vessel, so that for different atoms we may ignore the dependence of the phase of the radio-frequency field on position and take account of the time variation only;

(iii) it decays at time t later than t_0. We need make no distinction between the time of emission of light and the time of arrival at the photo-detector since the time of flight is negligibly small compared with the lifetime of the excited atoms and with the period of the radio-frequency field. However, since the atoms are distributed at random in the experimental cell, there can be no interference between light from different atoms. The net photo-electric signal is given by summing the effects of single atoms;

(iv) to describe a steady state experiment the signal at time t is found by summing over random times of excitation t_0. That is, if the rate of excitation is constant, R, the number of atoms excited in the interval dt_0 is $R dt_0$ and the net effect is found by integrating over t_0. (This approach neglects the statistical fluctuations).

The problem is reduced, therefore, to calculating the light emitted at time t by an atom known to have been excited to a particular state (or superposition-state) at time t_0. The calculation of the last section has enabled us to calculate the state function at time t and we need now to write an expression of the intensity of the emitted light. We shall see in a later section that it is of interest also to consider the state vector of the atom after the emission of light.

4.1. EMISSION OF LIGHT

In the semi-classical theory applied to emission from atoms in energy eigenstates, the well-known procedure is to suppose that each excited atom will radiate like a set of independent oscillators whose dipole moments are given by the matrix elements of the electric dipole operator between a specified excited state and all lower states which connect with it. We need to generalize this procedure for the present situation where the atoms are being subjected to a time-dependent perturbation (the r–f field). We must now take dipole matrix elements between eigenstates of the perturbed system, and if we express these in terms of the energy eigenstates of the system without the radio-frequency field we shall find superposition-states and time-dependent phase factors both in the excited states and in the ground states. Moreover, the levels concerned are those of the frequency diagram rather than the time-independent term diagram. But we should notice that the transition amplitudes are very small if the system is far from resonance, (e.g. if the angle β is small in eqns (23, 24, 25) the amplitude a_+ remains very close to unity and the amplitude a_- is always extremely small).

We generally meet one of the following two cases:

(i) the ground state is single ($J = 0$), in which case the question of radio-frequency mixing does not arise;

or (ii) if the ground state has structure the intervals are generally so different from the intervals between the excited states that when the radio-frequency field is near resonance for the latter, it is far from resonance for the former.

The conclusion is (so long as either (i) or (ii) above is true) that the required electric dipole matrix element is between a superposition-state for the excited states and an energy eigenstate of the unperturbed system for the lower state.

a. Example

Let the two-level system which we formerly discussed now represent the excited state of an atom. We shall use m or m' to label the two eigenstates. We may use the equations of Section III 3.1.a for this case, provided we take account of radiative damping. This we do by multiplying the amplitudes a_+ and a_- of the eqn (24) by $\exp -\frac{1}{2}\Gamma(t-t_0)$, supposing that both states decay at the same rate.

Suppose that the atom in its ground state is also a two-level system labelled by μ, μ'. Then the electric dipole matrix elements which we need are $\langle\mu|\mathbf{P}|t\rangle$, where μ can be $+$ or $-$. The component of the radiation which passes an analyser parallel to the unit vector $\mathbf{e}$ depends on $\langle\mu|\mathbf{e}\,.\,\mathbf{P}|t\rangle$, and the intensity of light passing this analyser will be proportional to the modulus squared.

This will be the intensity at time t emitted by an atom excited at time t_0. We thus have

$$I(t, t_0) = K \sum_{\mu} |\langle \mu | \mathbf{e} \cdot \mathbf{P} | t \rangle|^2, \tag{28}$$

where K is an unimportant constant. (In the quantum field theory, this equation gives the probability of finding a photon at time t).

Using the expansion

$$|t\rangle = a_+(t)\,|+\rangle + a_-(t)\,|-\rangle, \tag{29}$$

with a_+, a_- given by (24) we have

$$I(t, t_0) = K \sum_{\substack{\mu \\ m, m'}} \langle \mu | \mathbf{e} \cdot \mathbf{P} | m \rangle \langle m' | \mathbf{e}^* \cdot \mathbf{P} | \mu \rangle \, a_m \, a_{m'}^* \tag{30}$$

The intensity at time t resulting from excitation at the uniform rate R is given by

$$I(t) = KRA_{mm'} \int_0^t a_m(t, t_0)\, a_{m'}^*(t, t_0) \exp -\Gamma(t - t_0) \mathrm{d}t_0 \tag{31}$$

$$= KR\, Tr(A\sigma). \tag{31a}$$

In writing eqn (31a) we regard A as a matrix whose (m, m') element is given by

$$A_{mm'} = \sum_{\mu} \langle \mu | \mathbf{e} \cdot \mathbf{P} | m \rangle \langle m' | \mathbf{e}^* \cdot \mathbf{P} | \mu \rangle, \tag{31b}$$

and σ, whose (m, m') component is given by the integral in eqn (31), as the steady state density matrix for the excited atoms.

The elements of σ are:

$$\sigma_{++} = \frac{1}{\Gamma} \left[1 - \tfrac{1}{2} \frac{b^2}{\Gamma^2 + p^2} \right] \tag{32}$$

$$\sigma_{--} = \frac{1}{\Gamma} \left[\tfrac{1}{2} \frac{b^2}{\Gamma^2 + p^2} \right] \tag{32a}$$

$$\sigma_{+-} = \sigma_{-+}^* = \frac{b}{2\Gamma} \frac{\delta + i\Gamma}{\Gamma^2 + p^2} \exp -i\omega_0 t, \tag{32b}$$

where $\mathrm{b} = \gamma H_1$, $\delta = \gamma(H - H_0)$, $p = \gamma[(H - H_0)^2 + H_1^2]^{\frac{1}{2}}$.

By suitable choice of the type and orientation of the analyser which transmits the light, the elements of the matrix A can be given real or imaginary values or zero. Thus the intensity of the light will be represented by a sum of elements of the matrix σ weighted by factors which are under the control of the experimenter. The diagonal elements of σ will contribute time-independent terms, but for those configurations of analyser which yield non-vanishing

values for the off-diagonal elements of A, we shall find in the sum terms of the form

$$\left(\frac{b\delta}{\Gamma^2+p^2}\right)\begin{matrix}\sin\\\cos\end{matrix}\omega_0 t \quad \text{and} \quad \left(\frac{b\Gamma}{\Gamma^2+p^2}\right)\begin{matrix}\sin\\\cos\end{matrix}\omega_0 t. \tag{33}$$

The intensity of the light will therefore be given as a sum of resonance functions of the Lorentzian type with denominators (Γ^2+p^2). The width at half intensity is $2[(\Gamma/\gamma)^2+H_1{}^2]^{\frac{1}{2}}$. Arising from the off-diagonal elements are terms modulated at the driving frequency. The amplitudes of modulation are resonance functions of the bell-shaped type, $\Gamma/(\Gamma^2+p^2)$, or the dispersion-shaped type, $\delta/(\Gamma^2+p^2)$. It is worth noticing that the terms which arise from the diagonal elements of σ are proportional to b^2 whereas those arising from the off-diagonal element are proportional to b.

The resonance functions which we have found are precisely those which arise in the Bloch theory of magnetic resonance. Their occurrence here and in that theory may be traced to the fact that the signal which is being described is to be associated with a multipole of order 1 in the expansion of the density operator; in other words they are characteristic of the polarization of the system. If we were to look into the detail of the type of polarizer required to bring out the off-diagonal elements of σ we should find that of necessity we must use a circular or elliptical analyser; a linear analyser would not suffice. Similarly, in order to excite the system as we have supposed at time t_0 into the state $|+\rangle$, that is to say, to excite a polarized system, we would have required a circular or elliptical polarizer.

Had we studied a spin-1 system it would have been possible to contemplate alignment as well as polarization. Alignment can be generated and monitored by linear polarizers and analysers. In this case the resonance functions are slightly more complicated. They are given by Dodd and Series.[67] The monitoring of the density matrix by various means has been discussed by Carver and Partridge.[68]

5. Modulation in Double Resonance and Optical Pumping Experiments

5.1 Modulation in fluorescent light

We have shown how the effect of a radio-frequency field on a system near resonance is by no means completely described in terms of its effect on the populations of the states. The field injects coherence into the atomic system, which may formally be described in terms of the off-diagonal elements of a density matrix. We have shown how this coherence might be expected to give

rise to modulation in the fluorescent light in a double resonance experiment.

These expectations have been fully borne out by experiment. Figures 33a to d show resonance curves for the amplitude of modulation of light in the Brossel–Bitter experiment.[69] Here we are monitoring a spin-1 system, so the

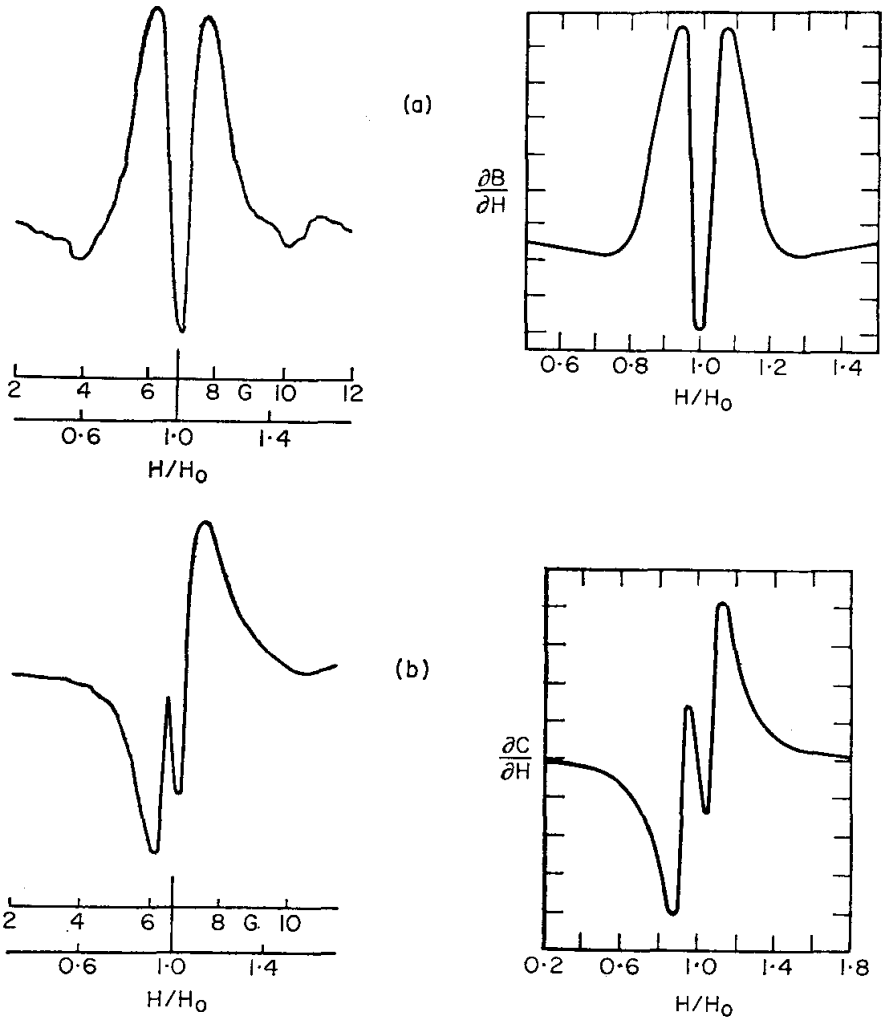


FIG. 33. Resonance functions for amplitude of modulation in the Brossel–Bitter experiment. a, b. In-phase and quadrature components at the fundamental frequency. The curves are derivatives with respect to magnetic field of the amplitude of modulation. Experimental curves on the left: theoretical curves on the right. (After Dodd, *et al.*[69])

curves are not the simple Lorentzians but the more complicated functions referred to in the last section. In fact, the curves were taken by imposing on the system a low frequency modulation and by phase-sensitive detection so that the curves are, in fact, derivatives of the resonance functions.

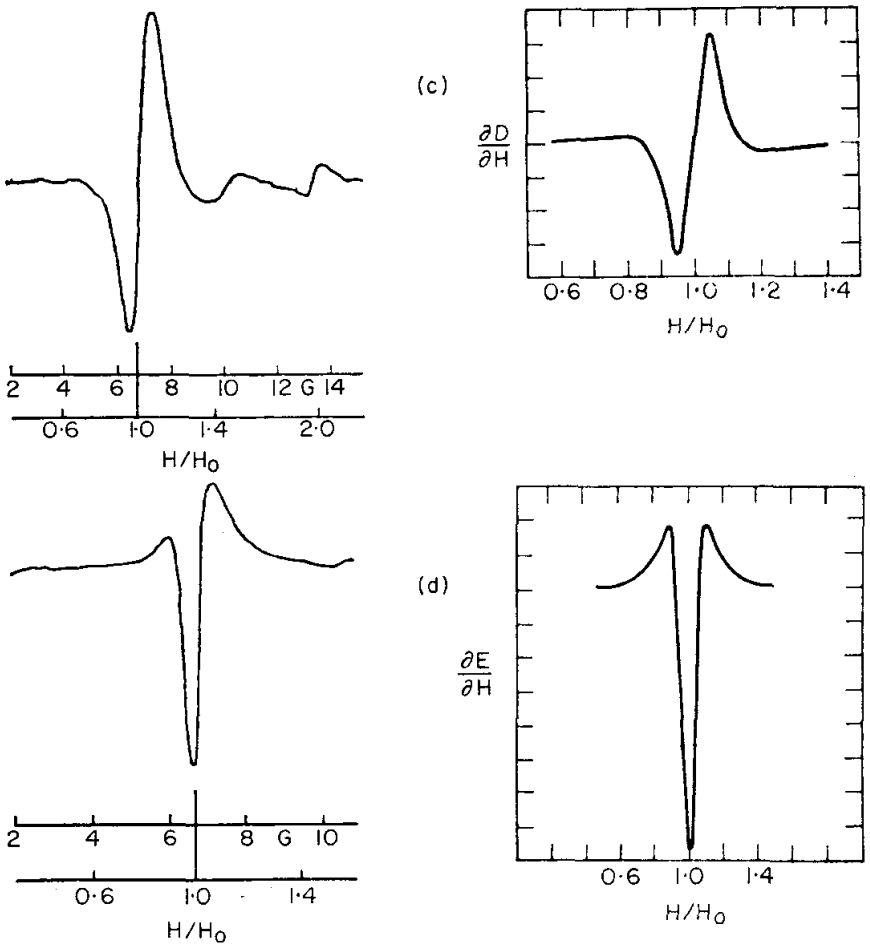


FIG. 33. c, d. In-phase and quadrature components at the second harmonic. (After Dodd *et al.*[69])

5.2. MODULATION IN ABSORPTION

Our calculation of the radio-frequency interaction holds also for atoms in the ground states (provided an inequality of population has been achieved by optical pumping). Just as coherence between the excited states is shown by modulation in the fluorescent light, so also one might expect coherence between ground states to be shown by modulation in absorption. This too has been studied experimentally. Figure 34 shows modulation in absorption from the metastable state 2^3S_1 in helium.[70]

6. TRANSFER OF COHERENCE

6.1. TRANSFER OF COHERENCE BY EXCITATION

Consider now a system in which coherence has been generated by a radio-frequency field, for example, the ground states in an optical pumping experi-

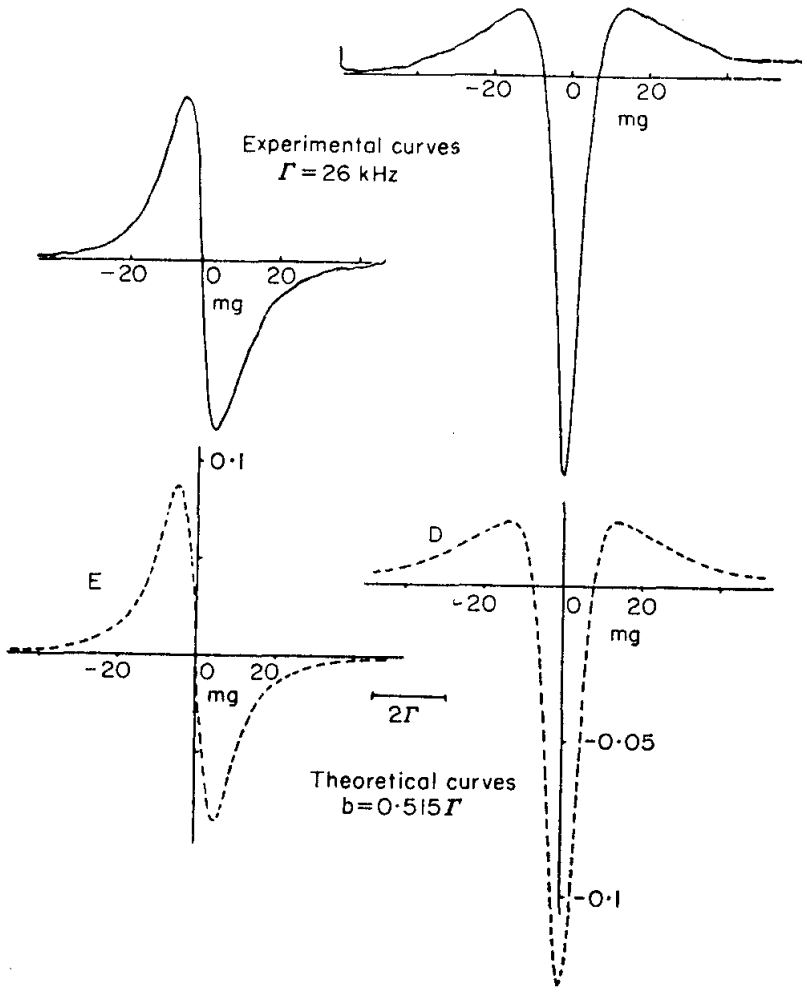


FIG. 34. Experimental and theoretical resonance curves for modulation in absorption from the metastable state 2^3S_1 in helium. In-phase and quadrature components at the second harmonic. These are to be compared with figures 33c and d. Experimental conditions: no polarizer, linear analyser, $\omega_0/2\pi = 0{\cdot}5$ kHz, $\Gamma = 26$ kHz, $b/\Gamma = 0{\cdot}515$.
(After Partridge and Series.[70])

ment as in the last section (2^3S_1 helium). Suppose, for preciseness, we consider the coherence between the states $|+1\rangle$ and $|-1\rangle$ and ask what would be the effect of illuminating the atom with light polarized parallel to the magnetic field (π-light; $\Delta m = 0$). There exist dipole matrix elements between 2^3S_1 and 2^3P_1; the states $|\pm 1\rangle$ of 2^3S_1 connect with $|\pm 1\rangle$ of 2^3P_1 respectively. Therefore, provided the spectrum of the light spans both these transitions in absorption, one might expect that the coherence between $|\pm 1\rangle$ in 2^3S_1 would be transferred to $|\pm 1\rangle$ in 2^3P_1, and this indeed takes place. (See Fig. 35.)

We need however, to consider this possibility a little more carefully. While it is true that the coherence can be transferred by a single pulse process, it is not necessarily the case that such coherence will be transferred in a steady-state experiment. We can think out this question by analogy with the theory

of electric circuits. The coherence we are trying to inject into the upper state is represented by a periodic function at a specified frequency. The upper state (described by its density matrix) is to be thought of as a receiver through which the coherence is to pass. We know it will pass if the applied frequency matches the tuned frequency of the receiver to within its bandwidth. In our analogy the tuned frequency of the receiver is given by the interval between the eigenstates $|\pm 1\rangle$ selected by the optical transitions. The bandwidth is given by the excited

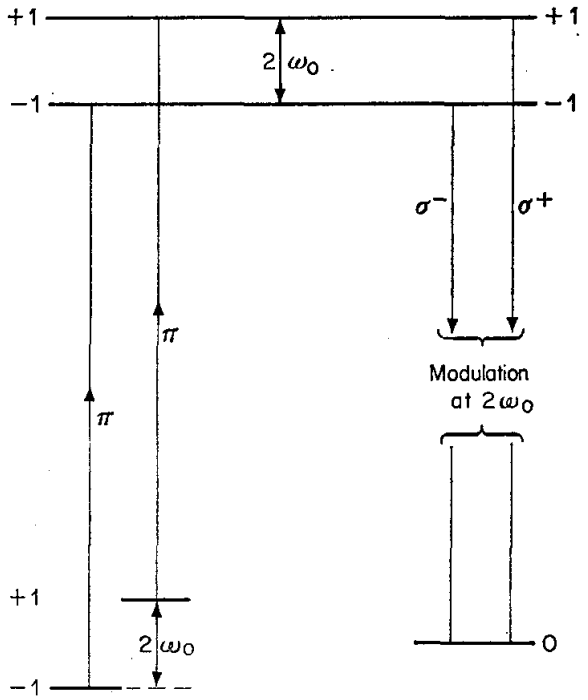


FIG. 35. Transfer of coherence from ground states to excited states and resulting modulation of the fluorescent light. The intervals between the levels are at the second harmonic of the driving frequency ω_0. They are not the same as the intervals in a static magnetic field.

state damping constant. In a practical case we might be trying to inject a frequency of some kilohertz or megahertz into excited states separated by an interval of a few megahertz but with a damping constant of several hundred megahertz. Because the bandwidth is so large the frequency mis-match is irrelevant in many actual cases. It is very easy to transfer coherence from the ground states to excited states in optical pumping experiments.

The coherence thus transferred is, of course, shown up in the fluorescent light. For this reason it is possible to monitor optical pumping signals both for modulated as well as unmodulated components by studying the fluorescent light.

6.2. TRANSFER OF COHERENCE IN DECAY

The possibility of transferring coherence exists for spontaneous decay as well as for excitation by light. We must consider two significant differences:

(i) spontaneous decay is an isotropic process whereas absorption can be anisotropic;

(ii) the restrictions imposed by the bandwidth of the receiver are very severe in the case when the receiving states are ground states.

Let us consider the consequences.

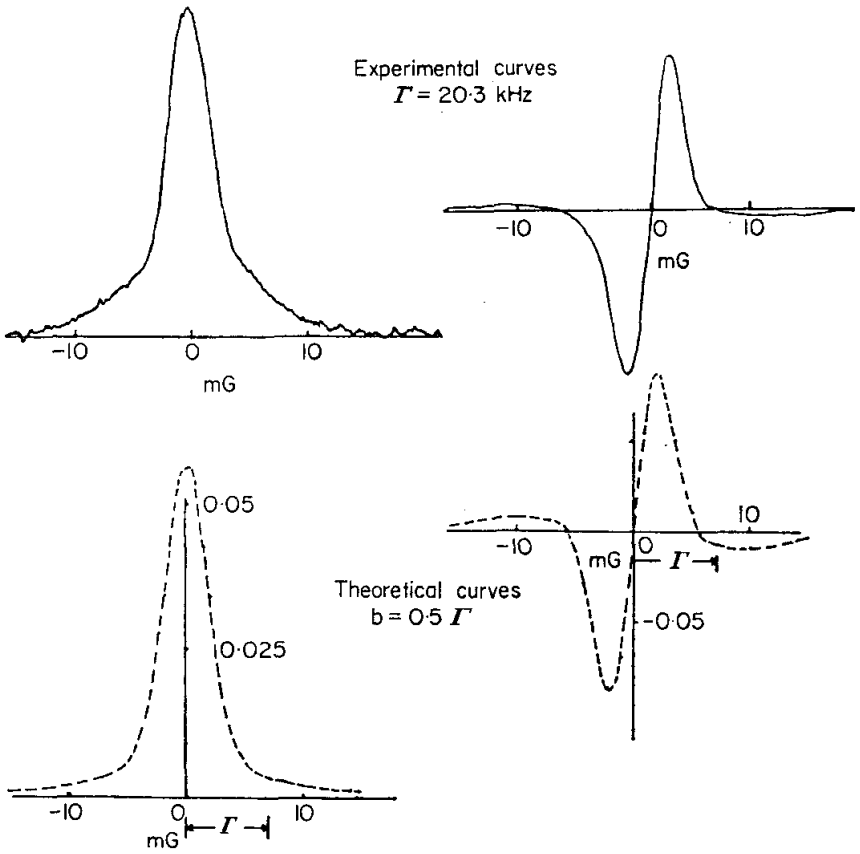


FIG. 36. Demonstration of circulation of coherence in optical pumping in helium. Experimental recordings compared with theoretical curves of the amplitude of modulation at $4\omega_0$. Experimental conditions: linear polarizer, no analyser, $\omega_0/2\pi = 0{\cdot}5$ kHz, $\Gamma = 20{\cdot}3$ kHz. $b/\Gamma = 0{\cdot}5$. (After Partridge and Series.[70])

(i) Although spontaneous emission is isotropic we may still resolve the perturbation into modes of sharp frequency and specified polarization. The integration over frequencies gives the result for a steady-state broad-band process which we analysed in the last section. It remains to perform the addition over the three orthogonal modes of polarization. This eliminates the possibility of transfer of coherence through the agency of different modes, (e.g. a

π–transition cannot couple with a σ^+ transition), but there remains the possibility for transfer by the same mode acting in both transitions (π with π, σ^+ with σ^+ and σ^- with σ^-).

(ii) Turning now to the bandwidth of the receiving states, this may be of the order of a few kilohertz or even a few hertz. Therefore, the frequency to be injected must match the interval between the ground state energy levels to within that tolerance. The levels we are here concerned with are those appropriate to the system perturbed by the radio-frequency field, namely, those which appear in the frequency diagram, rather than the energy levels appropriate to the system in a static field alone. The condition of matching finds mathematical expression in factors such as $\{\Gamma_\mu + i\omega_0[(\mu_1 - \mu_1') - (\mu_2 - \mu_2')]\}^{-1}$ which govern the probability for transfer of coherence from the (μ_1, μ_1') component of the density matrix to the (μ_2, μ_2') component via absorption followed by spontaneous decay[71]. For $\omega_0 \geqslant \Gamma_\mu$, the probability is very small unless $[(\mu_1 - \mu_1') - (\mu_2 - \mu_2')]$ is zero.

In Fig. 36 are shown resonance curves for the amplitude of modulation at $4\omega_0$ in the optical pumping experiment in 2^3S_1 of ^{4}He we referred to earlier. Evidence from the line-shape proves that the signals are due to coherence which was injected into the ground state, carried into the excited state and

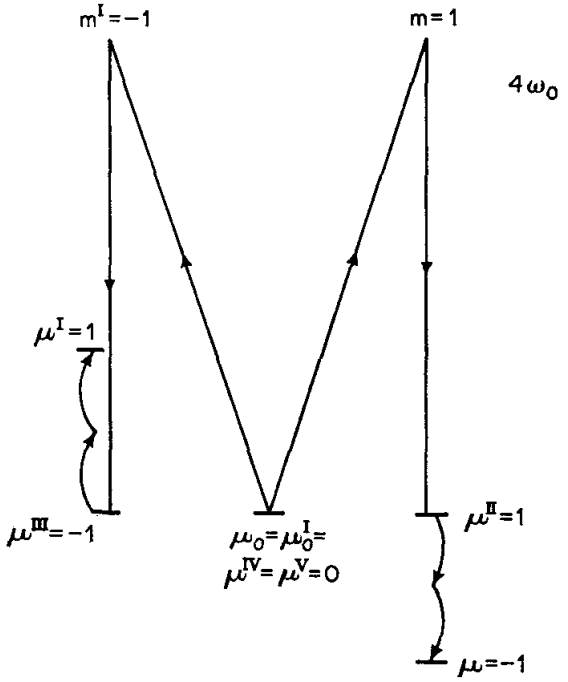


FIG. 37. Diagram showing the generation of coherence at $4\omega_0$ by the following sequence of processes:

(i) coherent absorption of σ^+ and σ^- light,
(ii) transfer of coherence by π mode of decay,
(iii) radio-frequency mixing.

back to the ground state by absorption followed by spontaneous decay, and monitored in the ground state by another absorption process. One of the sequences of transitions which contributed to this signal is represented in Fig. 37. It was shown that the strength of the signal was indeed frequency-dependent as is predicted by denominators of the form indicated in the last paragraph.

6.3. TRANSFER OF COHERENCE BY COLLISION

We have hitherto been speaking of processes taking place in individual atoms. We now consider the possibility of transferring coherence from one atom to another in collisions.

The process of spin exchange which we mentioned in an earlier section (I 6.9) may be represented as a transfer of longitudinal polarization by collision. The transfer of transverse polarization may be described as the transfer of coherence. There is no objection of principle, therefore, to the notion of the transfer of coherence by collisions. In fact, it takes place subject to the general rule that the frequency mis-match between the driving and receiving systems be not greatly in excess of the damping constant of the receiver.

An example illustrates what, at first sight, might appear to be surprising features of the phenomenon.

In a previous section we discussed the polarization of ^{3}He in the ground state 1^1S_0 by collisions with optically pumped ^{3}He atoms in the metastable state 2^3S_1. Magnetic resonance of the ^{3}He nuclei is monitored by detecting changes in the light absorbed by the metastable atoms. If one studies the absorption with the appropriate geometrical arrangement one finds modulation at the frequency at which one is driving the nuclear resonance. This is evidence that coherence injected into ground states has been transferred to metastable states of the atoms by collisions. The resonance curve (Fig. 23a) has a line-width of the order of a few hertz. It is to be noticed that such curves are obtained by measuring the absorption of light from atoms whose lifetime is of the order of milliseconds.

7. APPENDIX

We now return to the second case mentioned in Section III 3.

7.1. FIELD OSCILLATING SINUSOIDALLY PARALLEL TO A STATIC FIELD

The interaction Hamiltonian is

$$\begin{aligned}\mathscr{H} &= -\boldsymbol{\mu} \,.\, \mathbf{k}(H + H_1 \cos ft) \\ &= \gamma J_z(H + H_1 \cos ft). \end{aligned} \tag{34}$$

We assume that the field is not strong enough to break down the internal coupl-

ing (gyromagnetic ratio γ independent of the field). J_z is an operator. The Hamiltonian is readily diagonalized by choosing the z–axis for space quantization and it is possible to integrate the equation of motion directly, without first eliminating the time-dependence.

Thus:

$$i\hbar \frac{\partial}{\partial t}|t\rangle = \mathscr{H}|t\rangle \tag{35}$$

yields

$$|t\rangle = \left[\exp(-i/\hbar)\int_{t_0}^{t}\mathscr{H}(t')\mathrm{d}t'\right]|t_0\rangle = U(t, t_0)\,|t_0\rangle\,, \tag{36}$$

with

$$\int_{t_0}^{t}\mathscr{H}(t')\mathrm{d}t' = J_z[\gamma H(t-t_0)+(\gamma H_1/f)(\sin ft-\sin ft_0)]\,. \tag{37}$$

Use the expansion $\exp -iaJ_z \sin ft = \sum_{r=-\infty}^{\infty} J_r(aJ_z)\exp -irft$ (38)

(J_r is the Bessel function of order r).

Then

$$|m, t\rangle = \left\{\exp -im\omega(t-t_0)\left[\sum_{r=-\infty}^{\infty} J_r(am)\,\exp -irft\right]\right.$$
$$\left.\times\left[\sum_{s=-\infty}^{\infty} J_s(am)\exp -isft_0\right]\right\}|m, t_0\rangle \tag{39}$$
$$(\omega = \gamma H; \qquad a = \gamma H_1/f).$$

Rearranging, we have

$$|m, t\rangle = \left\{\sum_{r,\,s} J_r(am)\,J_s(am)[\exp -i(r-s)ft]\,[\exp -i(m\omega+sf)\,(t-t_0)]\right\}|m, t_0\rangle. \tag{40}$$

The periodic terms in the expansion are:

(i) the precessional term with frequency $(r-s)f$, phase-locked to the driving field, and

(ii) the nutational term with frequency $(m\omega+sf)$ associated with the time interval $(t-t_0)$.

For steady state excitation the phase of this nutational term, as before, is random. However, under certain conditions this random phase disappears from the density matrix, whose (m,m') component is:

$$\sigma_{mm'} = \sum_{\substack{r,\,s\\ r',\,s'}} J_r(am)\,J_s(am)\,J_{r'}(am')\,J_{s'}(am')\exp -i(r-s-r'+s')ft$$
$$\times \exp -i(m\omega+sf-m'\omega-s'f)\,(t-t_0)\,. \tag{41}$$

The condition for elimination of the random phase is:

$$(m-m')\omega = -(s-s')f,$$
$$\omega = (\text{integer}) \times f/(m-m'). \tag{42}$$

This is simply the condition for the degeneracy of certain levels in the frequency diagram. Degeneracy occurs when certain integral multiples of the driving frequency match multiples of the Larmor precessional frequency.

The frequency diagram for the case $J = 1$ is illustrated in Fig. 38. At $H = 0$ ($\omega = 0$) the levels $m = \pm 1$ are degenerate, but there exists a family of such degenerate pairs. The oscillating field splits each level into an infinite set labelled by integers r. Associated with each such level is a Bessel function whose order is the corresponding integer and whose argument is $am = m\gamma H_1/f$. The degeneracy at zero field is removed by the static field H.

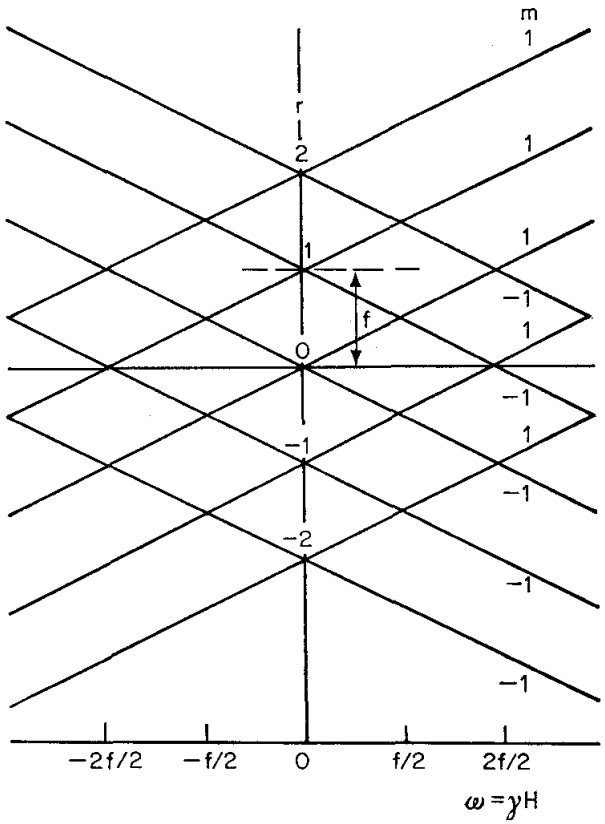


FIG. 38. Frequency diagram for system with $J=1$: magnetic field oscillating at frequency f parallel to static field H. Levels having $m=0$ are not shown.

The degeneracies at zero and at non-zero fields given by eqn (42) are a type of level-crossing similar to that discussed in a previous section. If account is taken of the actual width of excited levels the degeneracy extends over a region Γ. If atoms are excited from some initial state by light of polarization such that two different states m, m' can be excited, we find in the fluorescent

light an interference effect which we may describe as a resonance between the driving frequency and the Larmor frequency. These resonances have width Γ and suffer no radio-frequency broadening. The relative intensity of different resonances is given by a product of Bessel functions. The radio-frequency field strength governs these relative intensities. Resonances of this sort have been studied by Geneux and Favre,[72] Aleksandrov *et al.*[73] and by Chapman and Series.[74] It will be appreciated that modulation phenomena are associated with the resonances. Figure 39 shows an example of resonances in the 6^3P_1 state of mercury. The curves show the amplitude of modulation of the fluorescent light at the second harmonic of the driving frequency which in this case was 5 MHz. This is the interval between resonances, and was chosen so that the resonances were resolved against their width (2 MHz). The occurrence of the outermost resonances indicates that the Bessel function J_5 was significantly different from zero, which implies that γH_1 was a substantial fraction of 5 MHz.

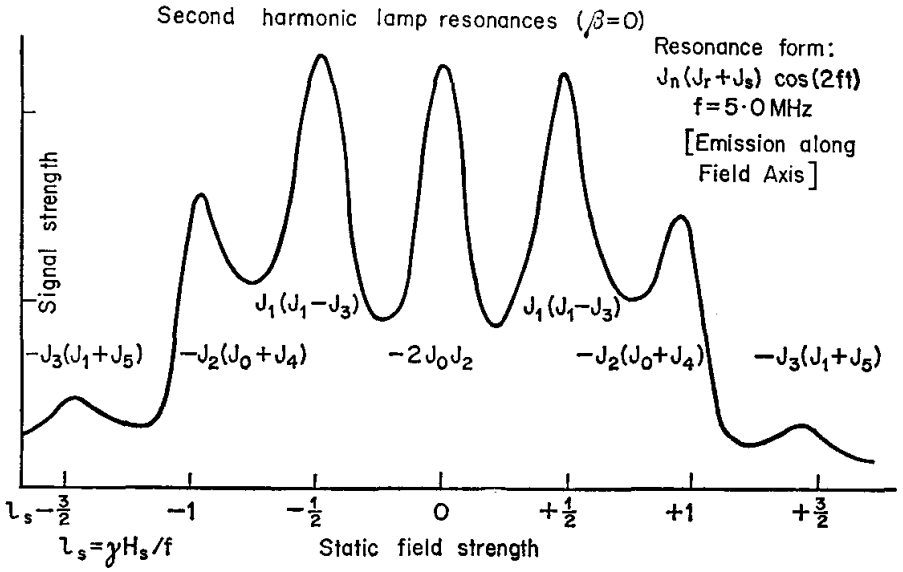


FIG. 39. Level-crossing in $^3P_1$ state of mercury subjected to parallel static and oscillating magnetic fields. Excitation by light polarized perpendicular to the fields.

IV RADIO-FREQUENCY INTERACTIONS

(b) PERTURBATIONS

We took as an example in the last section the case of a two-level system interacting with a rotating field. The resonance functions were symmetrical or antisymmetrical about the field $H = H_0$, which is the field where the applied frequency ω_0 exactly matches the interval between energy levels in Fig. 32a, i.e.,

the field where the levels cross in Fig. 32b. This is true also for multi-level systems where the levels are equally spaced as, for example, a spin-system having $J>\frac{1}{2}$, in a magnetic field. But it is not true for a rotating field interacting with a system where the levels are unequally spaced as, for example, in the case of a coupled-spin system which is being broken down by the magnetic field. In such a case the peaks of the resonances are displaced by an amount depending on H_1 and the resonance functions are no longer symmetrical. Such displacements are found, not only for the three-level system interacting with a rotating field, but also for a two-level system interacting with an oscillating field applied perpendicular to the static field. We shall consider these two cases.

1. System of Three Unequally Spaced Levels Interacting with Rotating Magnetic Field

Let us first apply the transformation which removes the time dependence and consider the energy levels in the rotating frame as a function of ω_0. Consider the example shown in Fig. 40a where the states are quantized with respect to the axis of rotation and the energy levels have been displaced from their positions in the laboratory frame by $m\omega_0$. The transverse field H_1 is supposed zero.

In Fig. 40(b) is shown the effect of applying the transverse field. The energy

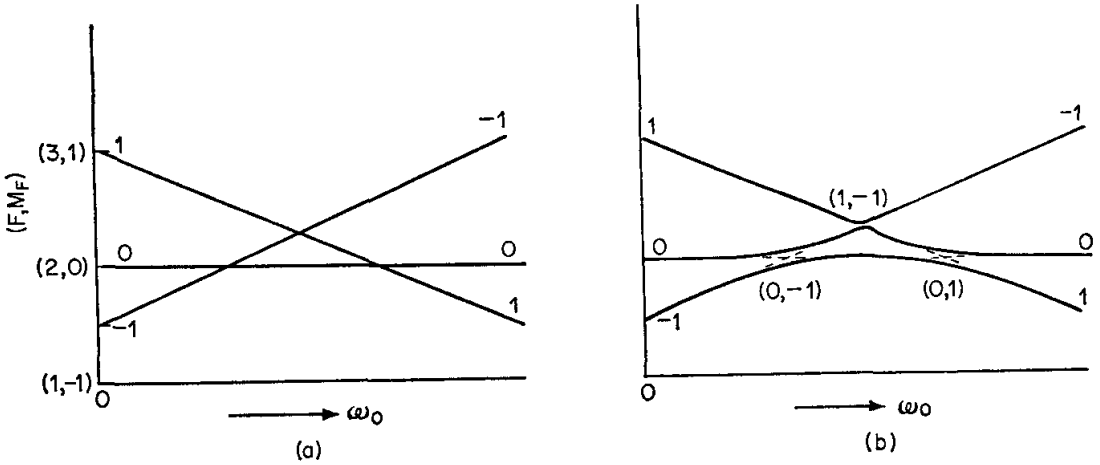


Fig. 40. System of three unequally spaced levels.
a. Energy in rotating frame, as function of ω_0.
b. Perturbation introduced by transverse field H_1.

levels are perturbed, and we must recall that the original quantum numbers are no longer good quantum numbers. The levels in this diagram are the eigenvalues of the transformed Hamiltonian $S\mathscr{H}'S^{-1}$ referred to in III 3.1 (iii).

1.1. Displacement of the Resonances

The centres of the resonances we shall take to be at the positions of closest

approach of the levels of the diagonalized Hamiltonian, though some argument is needed to justify this assumption.[75] These positions do not necessarily coincide with the intersections of levels in Fig. 40a. The intersection $(0,-1)$ for example is displaced because matrix elements of the perturbation exist between $|1\rangle$ and $|0\rangle$ but not between $|1\rangle$ and $|-1\rangle$.

As a first approximation to the correction we apply to the level 0 the perturbation it experiences from 1, Δ_{10}. The resonance condition is then given in terms of Δ_{10} and the Bohr frequencies Ω, as follows:

$$\Omega_0 - \Delta_{10} = \Omega_{-1} + \omega_0 . \tag{43}$$

Second order perturbation theory yields

$$\Delta_{10} = \frac{|b_{10}|^2}{(\Omega_1 - \omega_0) - \Omega_0} . \tag{44}$$

b_{10} is the matrix element of the perturbation. The denominator in (44) is the difference between the levels in Fig. 40a.

Since $|b_{10}|^2$ is proportional to ${H_1}^2$ the theory in this approximation predicts a displacement of the resonances proportional to ${H_1}^2$. This correction has been widely quoted in the literature (Salwen, for example).[76] It is customary to plot the position of the peak of the resonance against ${H_1}^2$ and to make a linear extrapolation to zero to obtain the unperturbed position. However, the approximation on which this procedure is based breaks down if Δ_{10} is comparable with the interval between the interacting levels. It is clear that a better solution is obtained by using for the energy denominator in eqn (44), the interval between the *perturbed* levels, Fig. 40b. An analytical solution has recently been given by Pegg who finds that in the case studied the displacement of the resonance was a function of $|H_1|$, not of ${H_1}^2$.

1.2. MULTIPLE QUANTUM TRANSITIONS

In the last section we concentrated on resonances corresponding to intersections of levels for which $\Delta m = \pm 1$, since the perturbation due to H_1 was supposed to have non-vanishing matrix elements between such states and no others. But the perturbation in second order connects levels with $\Delta m = \pm 2$, so that weaker resonances are to be expected for such intersections also. Thus a weaker resonance will occur for the intersection $(1,-1)$ in Fig. 40b. The characteristics of the resonance may be read from the figure:

(i) the position at which it occurs (the position of closest approach of the levels of the diagonalized Hamiltonian) will be approximately half-way between the intersections of $(0,-1)$ and $(1, 0)$ in Fig. 40(a);

(ii) the width of the resonance will be one-half that of the $\Delta m = \pm 1$ resonances because the levels intersect twice as steeply.

Such resonances are called multiple-quantum resonances because they correspond to an exchange of two radio-frequency quanta. Their occurrence in double resonance experiments was described in Section I 3.1.

2. Two Level System with Oscillating Field Perpendicular to Static Field

This case, though apparently quite different from the preceding case, nevertheless has much in common with it.

2.1. Displacement of the Resonances

Figure 41a shows the energy levels as a function of H in the laboratory frame. Resonances at $+H_0$ and $-H_0$ are induced by the clockwise and counter-clockwise rotating fields into which the oscillating field may be resolved. Each of these resonances perturbs the other so that their peaks do not fall exactly at the field $\pm H_0$. The shifts are known as Bloch–Siegert shifts. They may be understood by reference to the frequency diagram. (See Fig. 41b).

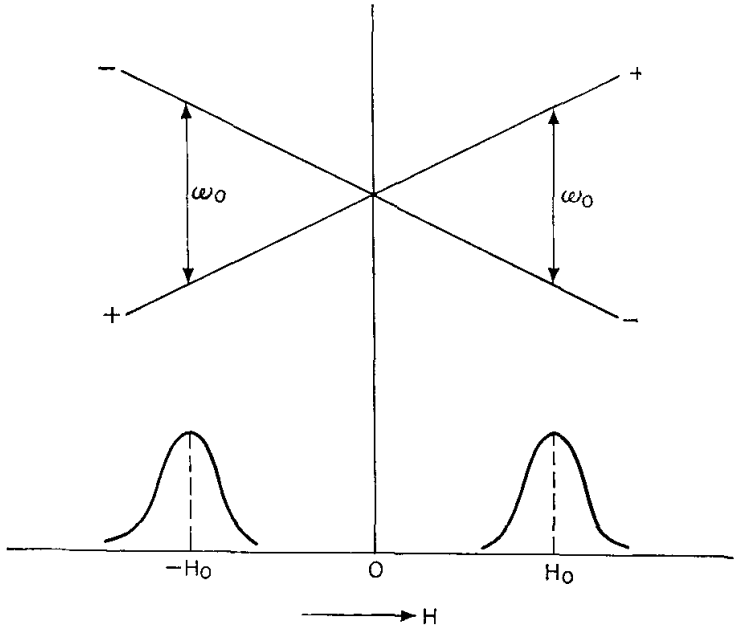


Fig. 41a. Resonances for spin-½ system at positive and negative values of the field.

Consider first that component of the field which gives rise to the resonance at $-H_0$. The frequency diagram, Fig. 41b, shows levels in the region of $+H_0$ separated by approximately the interval ω_0. The equations for the levels are:

$$\begin{aligned}\Omega_{mn} &= m(-\omega_0)+n[(\omega+\omega_0)^2+|b_{+-}|^2]^{\frac{1}{2}} \\ &\approx -m\omega_0+n(\omega+\omega_0)\,[1+\tfrac{1}{2}|b_{+-}|^2/(\omega+\omega_0)^2]\,, \qquad (45)\end{aligned}$$

where m, the quantum number along the static field, takes the values $\pm\frac{1}{2}$ as does n, the quantum number referred to $H_{\text{eff}} \cdot \omega = H$, and $b_{+-} = \frac{1}{2}\gamma H_1$ is the matrix element of the perturbation.

The resonance near H_0 is stimulated by the other component of the rotating field. This resonance occurs at that value of the field for which the interval between Ω_{++} and Ω_{--} is equal to ω_0, i.e. when

$$\omega_0 = \Omega_{++} - \Omega_{--} = -\omega_0 + (\omega_{\text{res}} + \omega_0)\,[1 + \tfrac{1}{2}|b_{+-}|^2/(\omega_{\text{res}} + \omega_0)^2],$$

where
$$\omega_{\text{res}} = \omega_0 - \tfrac{1}{2}|b_{+-}|^2/(\omega_{\text{res}} + \omega_0) = \gamma H_0 - \delta. \tag{46}$$

δ is the Bloch–Siegert shift.

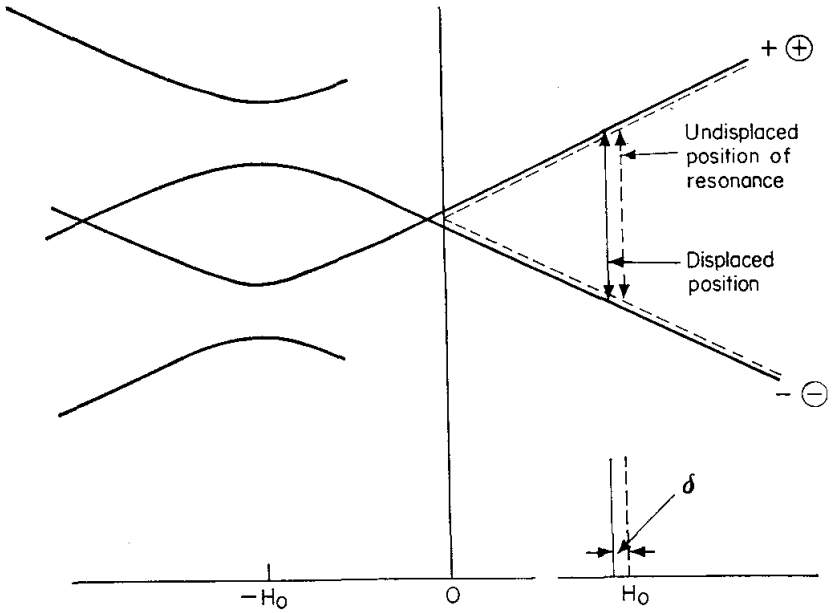


FIG. 41b. Bloch–Siegert shift illustrated by means of the frequency diagram.

2.2. MULTIPLE QUANTUM RESONANCES

Consider now the frequency diagram for the component of the field which stimulates the resonance at $+H_0$ (Fig. 42). We see the possibility of a resonance near $3H_0$, since here the pair of levels $-+$ and $+-$ (upper and lower, respectively) are separated by ω_0. Since the quantum numbers (laboratory frame) are inverted, this resonance needs the counter-rotating component of the field. The amplitudes of the interacting levels depend on the first rotating field, and are given by b_{+-}/p (eqn (24)). The transition matrix element is proportional to b_{+-}, so the amplitude of the resonance is proportional to $|b_{+-}|^2 b_{+-}$, and the transition probability to $|b_{+-}|^6$.

The position of the resonance is not exactly at $3H_0$ but is displaced by an amount equal to the Bloch–Siegert shift.

From Fig. 42 we may construct a further frequency diagram corresponding to the resonance near $3H_0$ by transforming to the counter-rotating frame

(Fig. 43a) and back to the laboratory frame (Fig. 43b). We now see the possibility of a resonance near $5H_0$ induced by the first rotating component. The process may be continued indefinitely.

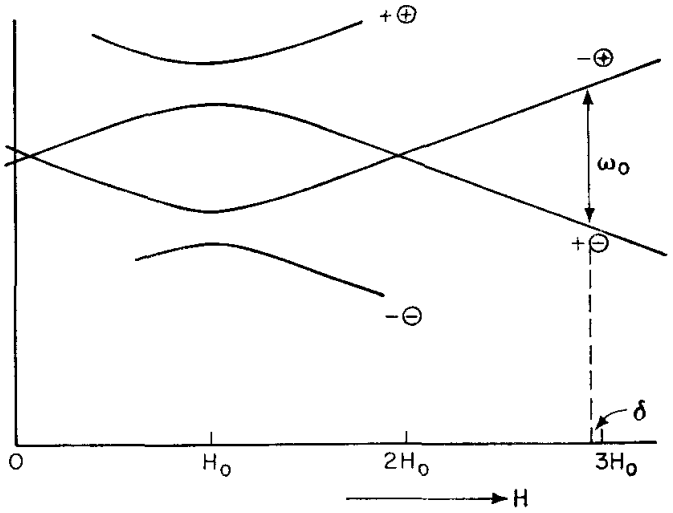


FIG. 42. Possibility of resonance near $3H_0$ illustrated by means of the frequency diagram.

Each new diagram contains two new levels. For each successive resonance a new factor $|b_{+-}|^4$ appears in the transition probability. If the resonances are studied with increasing values of b_{+-} (proportional to H_1) successive resonances appear in turn. The diagrams represent a treatment by successive approximation.

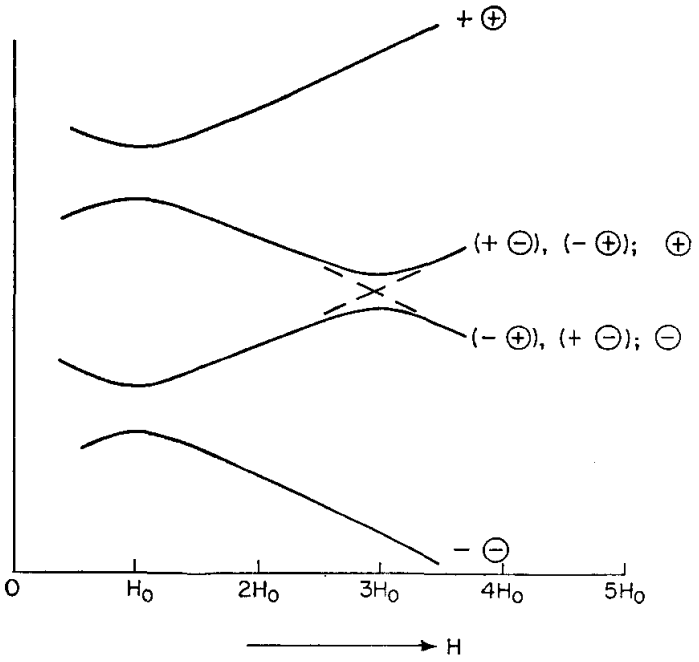


FIG. 43a. The frequency diagram of Fig. 42 transformed to the counter-rotating frame. The perturbation due to the transverse field H_1 (counter-rotating component) is shown.

A formal treatment which is equivalent to the foregoing approach has been given by Shirley [77] and has been adapted by Pegg[75] in the work previously quoted.

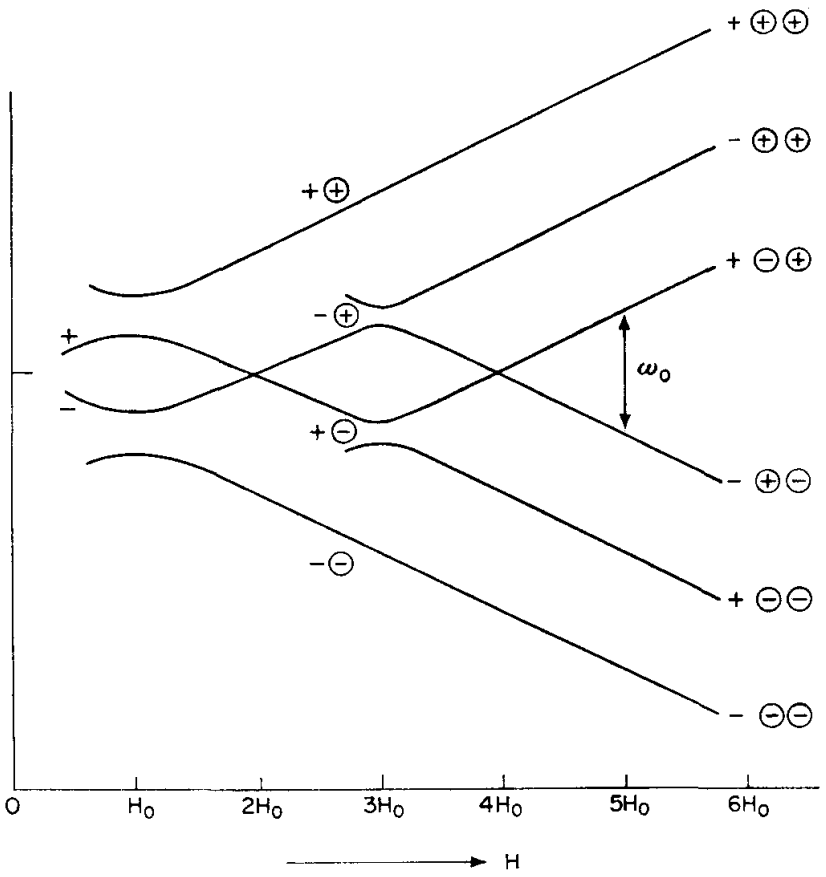


FIG. 43b. Frequency diagram in the laboratory frame representing the effects of rotating field and counter-rotating field in first approximation. The diagram shows the possibility of a resonance near $5H_0$.

3. Two Level System with Oscillating Field: Hanle Effect

Figures 41b and 42 show frequency diagrams for the two rotating fields separately, and in each case they are valid in the region $H = 0$. In Fig. 43b, representing the case of both fields acting together, the levels near $H = 0$ have not been drawn since, in this region, the perturbations due to both fields are comparable in strength and neither may be regarded as a perturbation on the other. The symmetry of the situation argues for a diagram which is symmetrical about $H = 0$, whereas both 41b and 42 are asymmetrical.

The situation near $H = 0$ may be analysed by regarding the static field as a perturbation on the oscillating field, treating the latter by the method given in Section III 7.

Consider the oscillating field $H_1 \cos ft$, together with the small, static trans-

verse field H acting on our two-level atomic system (Fig. 44). Choose an axis of quantization parallel to the oscillating field (Oz). Let Ox be the direction of the static field.

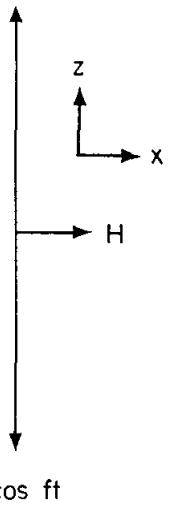


FIG. 44. Hanle effect with an oscillating field. The interaction with the transverse field H is regarded as a small perturbation on the interaction with the oscillating field $H_1 \cos ft$.

The Hamiltonian is

$$\begin{aligned}\mathscr{H} &= (\omega_1 \cos ft) J_z + \omega J_x \\ &= \mathscr{H}_0 + \mathscr{H}_1. \end{aligned} \tag{47}$$

The equation of motion under $\mathscr{H}_0$ above may be solved exactly to find the states $|t, t_0, \pm\rangle$ which develop at time t from the states $|\pm\rangle$ at t_0. We have

$$|t, t_0, +\rangle = \sum_r J_r(\tfrac{1}{2}a) \exp -irft \sum_s J_s(\tfrac{1}{2}a) \exp isft_0 \, |+\rangle \tag{48}$$

$$|t, t_0, -\rangle = \sum_{r'} J_{r'}(\tfrac{1}{2}a) \exp ir'ft \sum_{s'} J_{s'}(\tfrac{1}{2}a) \exp -is'ft_0 \, |-\rangle. \tag{49}$$

The following is also a solution:

$$|t, t_0, -\rangle^* = \sum_{r'} J_{r'}(\tfrac{1}{2}a) \exp ir'ft \sum_{s'} J_{s'}(\tfrac{1}{2}a) \exp is'ft_0 \, |-\rangle. \tag{50}$$

This state, which develops from

$$|t_0, t_0, -\rangle^* = \sum_q J_q(a) \exp iqft_0 \, |-\rangle. \tag{51}$$

at time t_0 differs from $|t, t_0, -\rangle$ by an arbitrary phase factor. We shall find $|t, t_0, -\rangle^*$ more convenient than $|t, t_0, -\rangle$ because its phase is more conveniently related to that of $|t, t_0, +\rangle$ than is the phase of $|t, t_0, -\rangle$.

Notice that linear superpositions of the states we have written down are also solutions of the equation of motion under $\mathscr{H}_0$.

The complete problem requires the addition of $\mathscr{H}_1$ to $\mathscr{H}_0$, so we try to find linear superpositions which are also eigenstates of $\mathscr{H}_1$.

We notice that there are degeneracies in the t-dependence of $|t, t_0, +\rangle$ and $|t, t_0, -\rangle^*$, namely, when $r' = -r$.

Now consider the superposition states:

$$|t, t_0, \pm\rangle^x = (1/\sqrt{2})\,[|t, t_0, +\rangle \pm |t, t_0, -\rangle^*]$$
$$= (1/\sqrt{2}) \sum_s J_s(\tfrac{1}{2}a) \exp isft_0 \left[\sum_r J_r(\tfrac{1}{2}a) \exp -irft|+\rangle \right.$$
$$\left. \pm \sum_{r'} J_{r'}(\tfrac{1}{2}a) \exp ir'ft|-\rangle \right]. \tag{52}$$

For those terms for which $r' = -r$ we can take out the common factor $J_r(\frac{1}{2}a) \exp -irft$, using $J_r(\frac{1}{2}a) = (-1)^r J_{-r}(\frac{1}{2}a)$.

We obtain

$$|t, t_0, +\rangle^x = (1/\sqrt{2}) \sum_s J_s(\tfrac{1}{2}a) \exp isft_0 \left\{ \sum_{r \text{ even}} J_r(\tfrac{1}{2}a) \exp -irft \right.$$
$$\left. \times [|+\rangle + |-\rangle] + \sum_{r \text{ odd}} J_r(\tfrac{1}{2}a) \exp -irft\,[|+\rangle - |-\rangle] \right\} + ,\ldots \tag{53}$$

and similarly for $|t, t_0, -\rangle^x$.

The states $(1/\sqrt{2})\,[|+\rangle \pm |-\rangle]$ are eigenstates of $\mathscr{H}_1$ with eigenvalues $\pm\frac{1}{2}\hbar\omega$. Thus the states $|t, t_0, \pm\rangle^x$ have the following properties:

(i) they satisfy the wave equation under $\mathscr{H}_0$;

(ii) with neglect of terms $r' \neq -r$, the even and odd parts of the wave function are separately eigenfunctions of $\mathscr{H}_1$ for all t, since

$$\mathscr{H}_1[|+\rangle \pm |-\rangle]/\sqrt{2} = \pm\tfrac{1}{2}\hbar\omega\,[|+\rangle \pm |-\rangle]/\sqrt{2}.$$

Thus the states $|t, t_0, \pm\rangle^x$ are convenient zero-order states on which to base a small perturbation $\mathscr{H}_1$.

Let us calculate the matrix of $\mathscr{H}_1$ in this basis. For the diagonal elements we have:

$${}^x\langle t, t_0, \pm|\mathscr{H}_1|t, t_0, \pm\rangle^x$$
$$= \pm\tfrac{1}{4}\hbar\omega \left[\sum_r J_r(\tfrac{1}{2}a) \exp -irft \sum_{r'} J_{r'}(\tfrac{1}{2}a) \exp -ir'ft \right]$$
$$\times \left[\sum_s J_s(\tfrac{1}{2}a) \exp isft_0 \sum_{s'} J_{s'}(\tfrac{1}{2}a) \exp -is'ft_0 \right] + c.c.$$
$$= \pm\tfrac{1}{4}\hbar\omega \sum_p J_p(a) \exp -ipft + c.c.$$
$$= \pm\tfrac{1}{2}\hbar\omega \sum_p J_p(a) \cos pft \tag{54}$$

(We have used a Bessel function contraction theorem, with $p = r+r'$; by the same theorem the second square bracket is identically unity.)

$$= \pm\tfrac{1}{2}\hbar\omega\left[J_0(a) + \sum_{\substack{p=2 \\ \text{even integers}}}^{\infty} 2J_p(a)\cos pft\right] \tag{54a}$$

For the off-diagonal elements,

$$\begin{aligned} {}^x\langle t, t_0, \mp|\mathscr{H}_1|t, t_0, \pm\rangle^x &= \pm\tfrac{1}{4}\hbar\omega \sum_p J_p(a)\exp -ipft - c.c. \\ &= \pm\tfrac{1}{2}i\hbar\omega \sum_p J_p(a)\sin pft \\ &= \pm\tfrac{1}{2}i\hbar\omega\left[\sum_{\substack{p=1 \\ \text{odd integers}}}^{\infty} 2J_p(a)\sin pft\right]. \end{aligned} \tag{54b}$$

Thus, for the time-independent component, and for the even harmonics, the matrix is diagonal. For the odd harmonics the matrix is diagonalized by a rotation of $\pi/2$ about Oz. The diagonalization removes the factor i from (54b).

If the oscillating field were absent ($H_1 = 0$), the matrix would be diagonal with elements $E_0 = \pm\frac{1}{2}\hbar\omega$. In the presence of the oscillating field we find diagonal elements,

$$E_0\left[J_0(a) + \sum_{\substack{\text{even} \\ \text{harmonics}}} 2J_p(a)\cos pft\right], \tag{55}$$

and off-diagonal elements

$$E_0\left[\sum_{\substack{\text{odd} \\ \text{harmonics}}} 2J_p(a)\sin pft\right]. \tag{56}$$

Thus the oscillating field has the effect of modifying the g-value of the spin system. Measured in the x-direction the g-value is predicted to be

$$g_x = g_0\left[J_0(a) + \sum_{\substack{\text{even} \\ \text{harmonics}}} 2J_p(a)\cos pft\right], \tag{57}$$

and in the y-direction,

$$g_y = g_0\left[\sum_{\substack{\text{odd} \\ \text{harmonics}}} 2J_p(a)\sin pft\right]. \tag{58}$$

In the z-direction the g-value remains unchanged.

The results have been obtained by finding the matrix of the perturbation $\mathscr{H}_1$ in a representation based on the eigenstates of $\mathscr{H}_0$, and are valid only for $\omega \ll f$. A more elegant treatment has recently been given by Pegg and Series.[77a]

The time-independent part of this result was obtained by Cohen-Tannoudji and Haroche,[78] and has been verified experimentally by them. Their derivation was based on the notion of "dressed atoms"—they derived the stationary states of a closed system consisting of (atoms + radio-frequency field), and

used the coherent state formalism of Glauber[79] to form superposition-states representing the oscillating field. The experimental study was by means of a level-crossing experiment for ground states.

V. REFRACTIVE INDEX AS A MONITOR OF OPTICAL PUMPING

1. Interaction of Light with Individual Atoms

Our discussion of the interaction between light and atoms has been based on the interaction with individual atoms. Thus, in III, eqn (31a), we derive an expression for the intensity of fluorescent light:

$$I(t) \propto \mathrm{Tr}(A\sigma), \tag{59}$$

where A is a matrix whose (m, m') element is

$$A_{mm'} = \sum_{\mu} \langle \mu | \mathbf{e} \cdot \mathbf{P} | m \rangle \langle m' | \mathbf{e}^* \cdot \mathbf{P} | \mu \rangle \tag{60}$$

and σ is the density matrix of the assembly.

Equation (59) is obtained by summing the intensities of light from the different atoms in the assembly.

Equations similar to (59) have been used also for absorption.

We should not lose sight of the fact that contributions to the intensity, either in emission or in absorption, may arise from the off-diagonal elements of the density matrix as well as from the diagonal elements.

2. Coherence Between Radiation from Different Atoms

The basis of eqn (59) must be re-examined when there is a possibility of interference between the light from different atoms. Such a possibility arises in the case of forward-scattering. The classical theory of refractive index interprets the dispersion of velocity of light in a medium as due to interference in forward scattering. Moreover, absorption is intimately related to dispersion and we monitor optical pumping by measuring the amount of light absorbed when a beam of light traverses the gas. We need to consider therefore, whether a theory of absorption based on the propagation of light through a medium (theory of dispersion) would lead us back to eqn (59), and whether this approach might not disclose other topics of interest. We shall see that the notion of an optically pumped vapour as a macroscopic medium having an anisotropic, time-dependent, complex refractive index is indeed useful.

2.1 Absorpton, Refractive Index, Electric Polarizability

The absorption coefficient of a medium is related to the imaginary part of the refractive index $n^{(i)}$. This may be obtained from the bulk polarization and this,

in turn, from the polarizability of individual atoms. We start from an expression for the polarizability tensor, $\alpha_{rs}^{(j)}$ of the jth atom, where r and s refer to the base vectors.

$$\mathbf{e}_{\pm 1} = \mp(\mathbf{i} \pm i\mathbf{j})/2^{\frac{1}{2}}, \qquad \mathbf{e}_0 = \mathbf{k}. \tag{61}$$

(**i**, **j**, **k**, are Cartesian unit vectors).

We have:

$$\alpha_{rs}^{(j)}(k, t) = \sum_{\substack{\mu \\ m=\mu+r \\ \mu'=m-s}} \frac{A_{rs}(\mu', \mu; m)}{\hbar(k_m - k_\mu - k - \frac{1}{2}i\Gamma_m)}\, a_\mu(t)\, a_{\mu'}{}^*(t) \ldots \tag{62}$$

where $$A_{rs}(\mu', \mu; m) = \langle \mu' | \mathbf{e}_s^* \,.\, \mathbf{P} | m \rangle \langle m | \mathbf{e}_r \,.\, \mathbf{P} | \mu \rangle,$$

k is the frequency of the incident light, $m, m' \ldots$ are excited states $\mu, \mu' \ldots$ are ground states, k_m, k_μ are Bohr frequencies, Γ_m is a damping constant. (Details of the calculation are to be found in Series.[80])

The bulk polarizability tensor, α_{rs}, is calculated by summing (62) over the N atoms per unit volume. These have a distribution of resonance frequencies $k_{m\mu} = k_m - k_\mu$ given by

$$\mathrm{d}N = (N/\Delta\pi^{\frac{1}{2}}) \exp\left[-\{(\delta k_{m\mu})^2/\Delta^2\}\right] \mathrm{d}k_{m\mu},$$

where Δ is a measure of the Doppler width of the distribution, and

$$\delta k_{m\mu} = k_{m\mu} - (k_{m\mu})_0, \text{ with } (k_{m\mu})_0 \text{ the peak.}$$

In making the sum we replace $a_\mu(t)\, a^*{}_{\mu'}(t)$ by its value $\sigma_{\mu\mu'}(t)$ averaged over atoms in ground states. The result is

$$\alpha_{rs}(k, t) = \sum_{\substack{\mu \\ m=\mu+r \\ \mu'=m-s}} \frac{NA_{rs}(\mu, \mu'; m)}{\hbar\Delta}\, \sigma_{\mu\mu'}(t)\, Z(x_{m\mu} + iy), \ldots \tag{63}$$

where $Z(x+iy)$ is the plasma dispersion function

$$Z(x+iy) = \pi^{-\frac{1}{2}} \int_{-\infty}^{\infty} \frac{\exp(-t^2)}{t-(x+iy)}\, \mathrm{d}t \;; \quad x_{m\mu} = \frac{k-(k_{m\mu})_0}{\Delta} \;; \quad y = \frac{\Gamma_m}{2\Delta}. \tag{64}$$

(We have ignored the contribution to refractive index which arises from excited atoms since, in optical pumping experiments, the bulk of the population is in ground states.)

a. Absorption

The absorption coefficient, χ, is related to the bulk polarizability, α, by

$$\chi_{rs} = (4\pi k/c)\alpha_{rs}^{(i)} \tag{65}$$

where $\alpha^{(i)}$ is the imaginary part of α. The imaginary part of α arises from the imaginary part of Z (eqn (63)).

Now, in exploring a resonance between ground states we are concerned with changes in the density matrix $\sigma_{\mu\mu'}$ which occur over relatively small varia-

tions of magnetic field, in the course of which Z remains practically constant. (The variation of Z is given by the variation of $x_{m\mu}$ with magnetic field, so we have to compare changes in the Zeeman splitting of the ground states, δk_μ, with the Doppler width, Δ).

If we can treat Z as constant, the absorption coefficient is proportional to

$$\sum_{\substack{\mu,\mu' \\ m}} A(\mu, \mu'; m)\, \sigma_{\mu\mu'}(t).$$

This is equivalent to eqns (59) and (60) (with m, m' and μ, μ' interchanged, because these equations refer to emission from excited states).

We have shown, therefore, that an analysis based on the classical theory of the propagation of light in dielectrics gives the same result for absorption, in optically thin vapours, as an analysis based on the interaction of light with individual particles.

b. Dispersion

We may expect that this approach would be rewarding if we considered the region away from absorption. We should then study the real part of eqn (63). The equation predicts that manifestations of optical pumping in ground states (resonances, modulation effects, etc.) could be studied by any technique which measured refractive index, since the optical pumping effects are described analytically by the density matrix, which is linearly related to the refractive index. Many such studies have indeed been made. The Faraday effect and magnetic birefringence, which measure various combinations of the tensor components of the anisotropic refractive index, have proved particularly useful.

For these dispersion studies it is advantageous to use monitoring light whose spectrum is displaced in frequency from the absorption region of the medium. This light does not then contribute to the optical pumping, and the effect of the pumping beam itself can be studied free from perturbations due to the monitoring.

2.2. EMISSION: RESONANCES IN EXCITED STATES

Our discussion has been based on the constancy of the function Z over the region of magnetic resonance. While it is true that this is constant to a high degree of approximation for ground state resonances, Z is not constant for excited state resonances. These resonances appear in eqn (63) as changes in the value of the function Z itself, since it is only through changes in $(k_{m\mu})_0$ that the effect of the magnetic field on excited states shows itself. ($(k_{m\mu})_0$ is simply the frequency of the (μ, m) Zeeman component of the absorption line). Now, changes of $(k_{m\mu})_0$ are significant only if they are comparable with the Doppler width. A detailed study of the consequences of this shows that curves of magnetic resonance in excited states, as explored by techniques which measure refractive index, are Doppler-broadened. This is in sharp contrast

with such curves as ordinarily studied (e.g. in the Brossel–Bitter experiment), where the great advantage of the technique is the elimination of Doppler broadening.

The difference between our present analysis and the earlier study of the Brossel–Bitter experiment is that we are now, for the first time, taking account of the possibility of coherence between light radiated by different atoms. In the Brossel–Bitter experiment, as in conventional experiments on resonance fluorescence, one studies the laterally-scattered light where there is no such coherence. In our present analysis of refractive index the coherence in forward-scattering makes itself felt by Doppler-broadening of the resonance curves.

There remains a further important point to be made in connection with forward-scattering. In optically thick vapours the transmitted light suffers multiple scattering and its intensity is not linearly related to the refractive index. A consequence of the multiple scattering is that the magnetic resonance curves experience coherence narrowing to a degree far in excess of coherence narrowing in the laterally scattered light. Indeed, zero-field level-crossing curves in sodium have been obtained by Hackett[81] which are actually narrower than curves obtained in lateral scattering. The analysis of this situation is complicated, and we shall not pursue it here.

The important points are:

(i) the result for emission is very different from that for absorption; and

(ii) coherence narrowing of excited state resonances is very marked in forward scattering.

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Coherence in Spontaneous Emission

H. HAKEN, R. HÜBNER AND K. ZEILE

In this paper we present a problem which was treated a couple of years ago in the lectures on quantum field theory given by Haken. The problem is quite simple. However, we present it here, because it shows how a fully quantum mechanical formulation of the field can be used to describe the coherence properties of the field in such a way as to reveal a close analogy with the classical treatment.

We begin by quickly repeating the Wigner–Weisskopf theory of spontaneous emission. We adopt the same notation as used in Section 4 of the chapter by Haken in this volume and split the total Hamiltonian which describes the single two-level atom, the field modes, and the interaction into two parts:

$$H = H_0 + H_I, \tag{1}$$

where

$$H_0 = \hbar v a_2^\dagger a_2 + \sum_\lambda \hbar\omega_\lambda b_\lambda^\dagger b_\lambda \tag{2}$$

and

$$H_I = \hbar \sum_\lambda (g_\lambda b_\lambda^\dagger a_1^\dagger a_2 + g_\lambda^* a_2^\dagger a_1 b_\lambda). \tag{3}$$

In H_I, non-resonant terms are as usual neglected. We make the following ansatz for the form of the wave function

$$\Phi(t) = A(t)\, a_2^\dagger \Phi_0 \exp(-ivt) + \sum_\lambda c_\lambda(t)\, b_\lambda^\dagger a_1^\dagger \Phi_0 \exp(-i\omega_\lambda t) \tag{4}$$

This is reasonable, since the total excitation energy is conserved in eqn (4). We insert (4) into the Schrödinger equation and compare the coefficients of the wave functions which describe the atom in its upper state or the atom in its lower state, and an additional light quantum. We then readily obtain the following equations

$$\left.\begin{aligned} \dot{A} &= \frac{1}{i}\sum_\lambda c_\lambda g_\lambda^* \exp[i(v-\omega_\lambda)t] \\ \dot{c}_\lambda &= \frac{1}{i} g_\lambda A \exp[i(\omega_\lambda - v)t] \end{aligned}\right\} \tag{5}$$

We use the hypothesis of Wigner–Weisskopf which assumes that the amplitude of the atom decays exponentially

$$A = \exp(-\gamma t) \tag{6}$$

and obtain from eqns (5)

$$c_\lambda = \frac{1}{i} g_\lambda \frac{1-\exp\left[-\gamma t + i\,(\omega_\lambda - \nu)t\right]}{\gamma - i\,(\omega_\lambda - \nu)} \tag{7}$$

and in a selfconsistent manner

$$\gamma = \sum_\lambda |g_\lambda|^2 \frac{\exp\left[\gamma t - i\,(\omega_\lambda - \nu)t\right] - 1}{\gamma - i\,(\omega_\lambda - \nu)}. \tag{8}$$

The right-hand side of eqn (8) can be evaluated for small values of γ, using the formula

$$\lim_{t\to\infty} \frac{1-\exp\left[i(\nu-\omega)t\right]}{\nu-\omega} = \frac{P}{\nu-\omega} - i\pi\delta\,(\nu-\omega) \tag{9}$$

and yields, in fact, a time independent quantity. So far we have merely reviewed the work of Wigner–Weisskopf.

Let us now interpret the results. In the Wigner–Weisskopf hypothesis, eqn (6), it is assumed that the upper state of the atom decays exponentially, but from this, it is hard to see how any coherence properties of the light field arise.

The conclusion usually drawn from the above solution is the following.

$$\frac{\mathrm{d}}{\mathrm{d}t} \int_{\substack{\Delta\omega \\ \Delta\Omega}} |c_\lambda|^2 \,\mathrm{d}\omega\,\mathrm{d}\Omega\,\delta\,(\nu-\omega) \tag{10}$$

represents the transition rate into photon states in a frequency interval $\Delta\omega$ over an angle $\Delta\Omega$, under conditions of energy conservation. Thus this quantity gives us a counting rate, but because it refers to photon numbers it says nothing about coherence properties. In order to find out what the coherence properties of the field are, we now apply the formalism of correlation functions. These have been extensively discussed in the chapter by Glauber in this volume. While Glauber discussed mainly the properties of correlation functions describing thermal (completely chaotic) light, we will now use this formalism for fields which are still coupled to their source. We first consider the following coherence function

$$\langle \Phi^+(0)\, E_x^{(+)}(\mathbf{x}, t)\, \Phi(0)\rangle, \tag{11}$$

where we use the decomposition

$$E_x^{(-)}(\mathbf{x}, t) = \exp(iHt)\left\{-\frac{i}{\sqrt{v}}\sum_{\lambda}\sqrt{(2\pi\hbar\omega_\lambda)}\,(\mathbf{e}_\lambda)_x \times \exp[-i\mathbf{k}_\lambda.\mathbf{x}]\,b_\lambda^{\dagger}\right\}\exp(-iHt). \tag{12}$$

H is the total Hamiltonian, (1), which acts on the creation operators $b_\lambda^{\dagger}$ and transforms them from the Schrödinger representation into the Heisenberg representation. $E^{(+)}$ is defined by the Hermitian conjugate of (12). In order to evaluate eqn (11), we merely have to calculate expressions of the form

$$\langle\Phi^{+}(0)\,b_{\lambda_0}(t)\,\Phi(0)\rangle \equiv \langle\Phi^{+}(0)\exp(iHt)\,b_{\lambda_0}\exp(-iHt)\,\Phi(0)\rangle. \tag{13}$$

This can be converted, by going into the Schrödinger representation, into

$$\langle\Phi^{+}(t)\,b_\lambda\,\Phi(t)\rangle. \tag{14}$$

When we insert the explicit wave function (4), we find that (13) vanishes, on account of the orthogonality of the unperturbed wave functions. At first sight one might conclude from this result that it implies that there is no coherence present at all. However, such a result could also be obtained in a completely classical theory if we averaged over all initial phases. Therefore we need the next order correlation function, in which the initial phase cancels out and which is defined by

$$\Gamma(t+\tau, t) = \langle\Phi^{+}(0)\,E^{(-)}(\mathbf{x}, t+\tau)\,E^{(+)}(\mathbf{x}, t)\,\Phi(0)\rangle. \tag{15}$$

Again we indicate briefly how to evaluate eqn (15). We insert eqn (12) and its Hermitian conjugate into (15). From the point of view of investigating the quantum mechanical aspects, we must evaluate expressions of the form

$$\langle\Phi^{+}(0)\,b_{\lambda_1}^{\dagger}(t+\tau)\,b_{\lambda_2}(t)\,\Phi(0)\rangle. \tag{16}$$

We again pass to the Schrödinger picture which leaves us with the expression

$$\langle\Phi^{+}(t+\tau)\,b_{\lambda_1}^{\dagger}\exp(-i\tau H)\,b_{\lambda_2}\,\Phi(t)\rangle. \tag{17}$$

Using the explicit form of the wave function (4) we find, for the last operator b_{λ_2} acting on $\Phi(t)$,

$$c_{\lambda_2}(t)\,a_1^{\dagger}\,\Phi_0\exp(-i\omega_{\lambda_2}t). \tag{18}$$

Now the operator $\exp(-i\tau H)$ yields just 1 when acting on (18), because there are no other transitions allowed. That part of (17) which stands on the left-hand side of the exponential operator can be evaluated in the same way as the right-hand side of (17), which led to (18). Using these results we find for (16) the following expression:

$$c_{\lambda_1}^*(t+\tau)\, c_{\lambda_2}(t) \exp\left[i\omega_{\lambda_1}(t+\tau) - i\omega_{\lambda_2} t\right]. \tag{19}$$

In order to evaluate (15) using this expression, we see that it is necessary to perform a double sum over λ_1, λ_2. This summation can be done using methods of ordinary classical physics, and we obtain finally

$$\Gamma(t+\tau, t) = 2\,\frac{v^4}{c^4} \exp\left[-2\gamma\left(t-\frac{r}{c}\right)\right] \frac{\sin^2\Theta}{r^2} |(\theta_{12})^2| \exp(-iv\tau) \exp(-\gamma\tau) \quad \text{for } ct-r>0$$

$$= 0 \quad \text{for } ct-r<0 \tag{20}$$

In (20) we have neglected γ^2 compared with v^2. θ_{12} is the atomic dipole moment matrix element. Θ is the angle between the atomic dipole moment and the direction of observation.

The amazing thing is that formula (20) could have been obtained equally well by using a completely classical treatment of the radiation field of a damped Hertzian oscillator. The result shows that the field E has an internal coherence time $1/\gamma$ which appears here, multiplying τ. Our result differs from one which is sometimes expressed, that coherence properties do not show up in the spontaneous emission of photons, because the photons have no phase. In fact, formula (10) does not contain a phase term. On the other hand, if measurements are performed which allow us to determine correlation functions of the form (15), we find exactly those coherence properties which we would expect from a classical treatment of a field with an internal coherence time $1/\gamma$.

We should add two warnings or remarks, however. Our result should not be interpreted as meaning that quantum electrodynamics is unnecessary and can be replaced by a fully classical theory. Finding one such analogy, or even several, is, of course, not sufficient, and quantum field theory could be dispensed with only if all relevant results could be explained in a classical theory. We would also want this new "classical" theory to be at least as simple in formulation as the old quantum electrodynamical theory. Our main conclusion from our above treatment is that quantum electrodynamics because of its inherent dualism between particle and wave aspects, is sufficiently versatile to account for finding photons in spontaneous emission, and also to account for the coherence properties of the fields in this process.

The second remark is that one might be tempted to interpret the result as follows: if one believes in the field as a real quantity, the coherence properties will automatically follow, since coherence is a property of the field itself. On the other hand, the correlation function (15) could be measured by photon counting, and the measurement would imply the interaction of light and matter. From this, one would conclude that only matter occurs in reality. We prefer to leave the discussion of this point open however, because the choice of the field as the real quantity has at least simplified the interpretation of our results.

Quantum Statistics of Optical Parametric Oscillation

R. GRAHAM

The subject of this paper is a rather old one; parametric oscillation was first discovered by Lord Rayleigh[1] in 1883. Nowadays the term has a wider usage. Any oscillation of a system with time-varying parameters is called a parametric oscillation. Here, however, I want to confine myself to the simplest case.

Let us consider two slightly damped harmonic oscillators (we call them "signal" and "idler" in the following) with frequencies ω_1 and ω_2, and damping constants κ_1 and κ_2. Let these two oscillators be coupled by some "parameter" and let us modulate this parameter with a harmonically varying force F_p of frequency ω_3. A simple calculation[2] then shows that, provided the matching condition

$$|\omega_3 - \omega_1 - \omega_2| \lesssim \kappa_1, \kappa_2 \tag{1}$$

is fulfilled, the effective damping in both oscillators is reduced, and above a certain threshold value of F_p self-sustained oscillations take place. Below threshold this device is called a parametric amplifier, above threshold we have the oscillator region. If the two harmonic oscillators of the system coincide, i.e. degenerate into a single oscillator, we call it degenerate parametric oscillation or subharmonic generation.

Examples of parametric oscillators in everyday life are, of course, numerous. An ordinary swing serves as an excellent example. If you sit on a swing and excite it by raising and lowering your centre of gravity at twice the swing frequency, you are then part of a degenerate parametric oscillator.

So far I have not mentioned a very important point, which is essential for parametric oscillation noise. You can sit on your swing and raise and lower your centre of gravity for a long time: nothing will happen if there is nobody nearby to give you the first push, or alternatively, if there is no other initial random excitation of the swing. In classical physics, one could imagine a situation in which there are no fluctuations. However, the situation changes completely if signal and idler are quantized. Vacuum fluctuations of these oscillators will always be present and these are sufficient for the build-

up of self-sustained oscillation. Thus, a quantized swing would easily be excited without the help of other initial excitations.

Therefore, to study the part which noise plays in parametric oscillation, it is helpful to set up an oscillator in a region where quantum effects should be measurable. This is at least partially achieved in the optical region.[3] In the optical region, signal and idler oscillations are provided by two modes of the electromagnetic field. (The idler mode may be some other elementary boson excitation, e.g. an optical or an acoustical phonon, in which case we have stimulated Raman and Brillouin scattering respectively). The coupling parameter is the susceptibility of the medium in which the electromagnetic modes propagate. If we chose a medium whose susceptibility depends linearly on the electric field strength E, we can modulate this coupling parameter by shining in a powerful beam of laser light of frequency ω_3. A simple experimental scheme is shown in Fig. 1. One of the first realizations of this scheme was achieved by Giordmaine and Miller.[4]

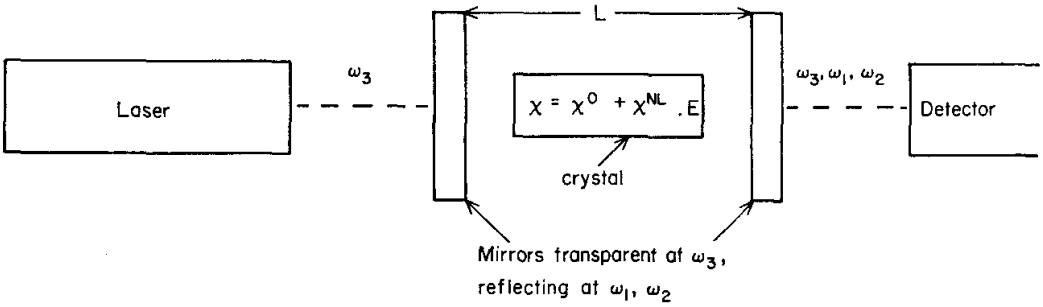


Fig. 1. Scheme of optical parametric oscillator (doubly resonant type).

Generation of optical signal and idler modes in this device is possible only if there are resonant cavity modes which fulfill the phase matching condition

$$| \mathbf{k}_3 - \mathbf{k}_1 - \mathbf{k}_2 | \leqslant \pi/L \tag{2}$$

for the wave vectors $\mathbf{k}_\lambda$ of the three modes involved. To achieve phase matching, it is necessary to compensate for dispersion in the crystal. This can be done by using a double refracting crystal and polarizing the pump in the extraordinary direction. It is then possible to choose ordinary signal and idler waves which satisfy both the energy and momentum matching conditions.[4]

Because of the central importance of noise in this problem, we wish to know the statistical properties of the signal and idler output of the parametric oscillator. The experimental answer to this question has not yet been given, because experimentally the parametric oscillator is difficult to control. In most cases, the output contains several clusters of signal/idler pairs,

because the mode selection of this device is rather poor.[4] However, the situation is improving, and there have recently appeared several papers on high efficiency parametric conversion.[5] Using these techniques together with a mode selecting device, it may be possible to perform the appropriate experiments. However in the following discussion I shall answer the question theoretically.

The Hamiltonian of the system shown in Fig. 1 may be written

$$H = \sum_{\lambda=1}^{3} \hbar\,\omega_\lambda\, b_\lambda^\dagger\, b_\lambda + H_{\text{cryst}} + H_{\text{cryst-field}} + H_{\text{pump}} + H_{\text{bath}} + H_{\text{bath-field}} + H_{\text{bath-cryst}}. \tag{3}$$

We have introduced here the creation and annihilation operators, $b^\dagger$ and b for the three modes in the cavity. The first term in the Hamiltonian describes the free motion of these modes. The free motion of the atoms of the crystal and their coupling to the field modes are described in the second and third terms. In the fourth term, the mode with frequency ω_3 is directly coupled to an external pump force F_p

$$H_{\text{pump}} = \frac{\hbar}{i}\,(F_p^*\exp(i\omega_3 t)\,b_3 - F_p\exp(-i\omega_3 t)\,b_3^\dagger). \tag{4}$$

There are losses in the system. The pump mode leaves the crystal without reflection, and the signal and idler modes are damped by mirror and diffraction losses. These losses are described by coupling the field to heat baths. The fifth and sixth terms in the Hamiltonian are introduced to take care of the free motion of the heat baths and their coupling to the field modes. The crystal itself is also coupled to its surroundings, and this is described in the last term.

In general the solution of this problem is very complicated, but Bloembergen has shown that the problem may be greatly reduced if we know which physical process is most important and dominant in the interaction of the field and the crystal.[6] In this case, although the field modes are coupled via the crystal, the crystal operators themselves can be completely eliminated from the problem. We are then left with three field modes, with slightly shifted frequencies $\bar{\omega}_\lambda$, interacting with each other via a nonlinear susceptibility, and slightly modified heat baths.[7] The effective Hamiltonian is

$$H_{\text{eff}} = \sum_\lambda \hbar\,\bar{\omega}_\lambda\, b_\lambda^\dagger b_\lambda + i\hbar\beta\,(b_1^\dagger\, b_2^\dagger\, b_3 - b_3^\dagger\, b_1\, b_2) + \frac{\hbar}{i}\,(F_p^*\exp(i\bar{\omega}_3 t)\,b_3 - F_p\exp(-i\bar{\omega}_3 t)\,b_3^\dagger) + H_{\text{bath}}^{\text{eff}} + H_{\text{bath-field}}^{\text{eff}} \tag{5}$$

Detuning effects are neglected for simplicity. β is a real coupling constant, and is proportional to the nonlinear susceptibility of the crystal.

Let me clarify the reasons why this effective Hamiltonian exists and under what circumstances. Very often in quantum field theory one has the task of eliminating some of the underlying field operators, in order to end up with an effective interaction between the remaining fields. However, it is not always possible to write down an effective Hamiltonian for this interaction, i.e. to achieve the elimination at least approximately by some unitary transformation. As a general rule, an effective Hamiltonian exists if only *virtual* excitations of the eliminated fields occur in the problem under consideration (compare for example, the elimination of the virtual meson excitations in nuclear force theory or of the virtual phonons in the theory of superconductivity). If, on the other hand, *real* transitions of the eliminated fields are involved in the problem, damping phenomena have to be taken into account to avoid divergences, and then the effective interaction cannot be described by an effective Hermitian Hamiltonian, because of the irreversibility of the damping phenomena. A good example of the latter case is the laser. Here, real electronic transitions (see Fig. 2a) are the origin of the nonlinear saturation and mode coupling effects. In fact, the elimination of the atomic variables[8] from the laser problem leads to equations of motion for the mode amplitudes alone (e.g. the van der Pol equation), which cannot be derived from an effective Hamiltonian.

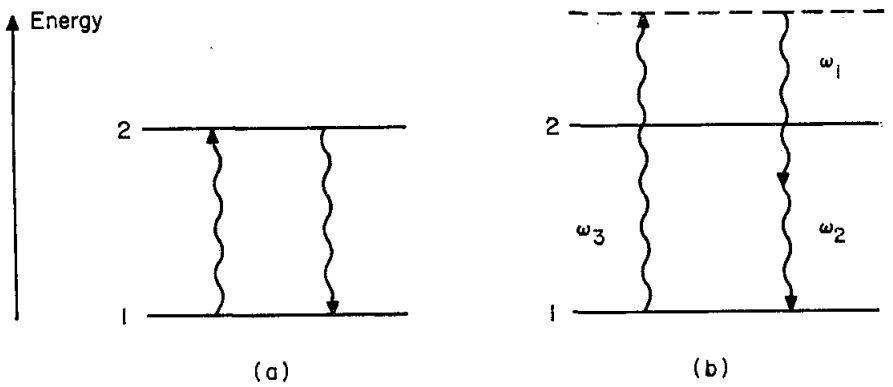


FIG. 2 a. Real electronic transitions in a two-level laser atom, causing mode coupling effects. b. Virtual electronic transitions in a two-level atom in a medium with nonlinear susceptibility, causing the parametric interaction between pump, signal and idler.

In the parametric oscillator case, we have mostly only virtual electronic transitions (Fig. 2b). This is why we can write down an effective Hamiltonian in this case (and in similar cases, for example, second harmonic generation or frequency conversion). If, however, one of the virtual transitions in Fig. 2b is tuned to resonance with the atomic transition, it becomes a real transition

and the effective Hamiltonian (5) is invalid. In this case only effective equations of motion for signal, idler, and pump can be written down, since there is no Hermitian Hamiltonian.[7] In the following, we treat only the nonresonant case shown in Fig. 2b, where (5) gives a complete description.

We may obtain some insight into the physics of the process by comparing the interaction part of the effective Hamiltonian

$$H_{\text{int}}^{\text{eff}} = i\hbar\beta(b_1^\dagger b_2^\dagger b_3 - b_1 b_2 b_3^\dagger) \tag{6}$$

with the interaction part of the two-level laser Hamiltonian

$$H_{\text{int}}^{\text{laser}} = g\sum_\mu (b\alpha_\mu^\dagger + b^\dagger \alpha_\mu). \tag{7}$$

Here $\alpha_\mu^\dagger$ is the raising operator for the μ-th two-level atom, α_μ the corresponding lowering operator. If we set up a correspondence between the signal and laser modes, i.e.

$$b_1 \leftrightarrow b; \qquad b_1^\dagger \leftrightarrow b^\dagger \tag{8}$$

then we also have the following operator correspondences

$$b_2^\dagger b_3 \leftrightarrow \sum_\mu \alpha_\mu; \qquad b_2 b_3^\dagger \leftrightarrow \sum_\mu \alpha_\mu^\dagger \tag{9}$$

$b_2^\dagger b_3$ describes the annihiliation of a photon in the pump mode with the simultaneous creation of a photon in the idler mode, thus corresponding to a transition from the higher energy level $\hbar\omega_3$ to the lower energy level $\hbar\omega_2$. The operator $\sum_\mu \alpha_\mu$ describes the corresponding process for electrons in the two-level laser atoms.

In the laser we have many independent electrons, pumped into the upper state of many independent two-level atoms. A light mode is created by stimulated transitions into the ground state. In the parametric oscillator we have many photons, pumped into a mode with energy $\hbar\omega_3$ which create a light mode (the signal) by stimulated transitions into a mode with lower energy $\hbar\omega_2$ (the idler). This process is called stimulated parametric emission. Noise in the laser system is caused by the atoms making spontaneous transitions, and similarly in the parametric oscillator system, noise is caused by spontaneous transitions of photons from the pump mode to the idler mode with the emission of a signal photon. This is called spontaneous parametric emission, and is induced by the vacuum fluctuations in the signal and idler.

In the limiting case of an infinite number of atoms in the laser, the correspondence of the interaction Hamiltonians goes over into equivalence, since

then the operator equation

$$\sum_{\mu=1}^{\infty} \alpha_\mu^\dagger = b_3^\dagger b_2 \tag{10}$$

satisfies all the properties of the operators on both sides of the equation. The other terms in the complete Hamiltonians are still different, so that the results differ in their finer details,[9] but the overall results for the parametric oscillator show a strong analogy with the laser results, even in the case of a finite number of laser active atoms.

Let us now start from the effective Hamiltonian and derive some more explicit results. First we derive a master equation for the density operator ρ of the three mode system, by eliminating the effective heat baths in the usual way.[10]

We obtain, in the interaction picture

$$\frac{\partial \rho}{\partial t} = \Big\{ \Big[\beta\,[b_1^\dagger b_2^\dagger b_3, \rho] + F_p\,[b_3^\dagger, \rho] + \sum_{\lambda=1}^{3} \left(\kappa_\lambda\,(\bar{n}_\lambda + 1)\,[b_\lambda \rho, b_\lambda^\dagger] + \kappa_\lambda\,\bar{n}_\lambda\,[b_\lambda^\dagger \rho, b_\lambda]\right) \Big] + [\text{Hermitian conjugate}] \Big\}. \tag{11}$$

κ_λ are the damping constants for the three modes, $\bar{n}_\lambda$ is the sum of the number of thermal quanta at frequency ω_λ and the number of quanta which are emitted spontaneously by the crystal atoms into this mode. At optical frequencies and realistic temperatures, $\bar{n}_\lambda$ is a very small number compared to 1.

The next step is to introduce a quasi-probability distribution function for the statistical operator. We could use the P-representation, the Wigner-function or the diagonal elements of ρ with respect to coherent states—all these quasi-distribution functions contain the same information as the statistical operator itself, and all multitime correlation functions may be easily calculated from them.[11] In the parametric oscillator case we expect very strong correlations between signal and idler. Consequently the P-representation does not always exist [12, 9] so instead we use a representation which does:

$$Q(\{\alpha_\lambda\}) = \frac{1}{\pi^3} \langle \{\alpha_\lambda\} \mid \rho \mid \{\alpha_\lambda\} \rangle, \tag{12}$$

where

$$b_\lambda \,|\, \{\alpha_\lambda\}\rangle = \alpha_\lambda \,|\, \{\alpha_\lambda\}\rangle. \tag{13}$$

Once we have calculated this function from the master equation, it can be used to calculate normally ordered multitime correlation functions.[11] All we have to do is to make the following substitutions

$$b_\lambda^\dagger \to \left(\alpha_\lambda^* + \frac{\partial}{\partial \alpha_\lambda}\right); \qquad b_\lambda \to \left(\alpha_\lambda + \frac{\partial}{\partial \alpha_\lambda^*}\right) \tag{14}$$

and to use Q as if it were a classical probability distribution.[11] For Q we obtain the Fokker–Planck type equation

$$\begin{aligned}\frac{\partial Q}{\partial t} = \Bigg\{ \Bigg[& \frac{\partial}{\partial \alpha_1} (\kappa_1 \alpha_1 - \beta \alpha_2^* \alpha_3) Q + \frac{\partial}{\partial \alpha_2} (\kappa_2 \alpha_2 - \beta \alpha_1^* \alpha_3) Q \\ & + \frac{\partial}{\partial \alpha_3} (\kappa_3 \alpha_3 + \beta \alpha_1 \alpha_2 - F_p) Q \\ & - \beta \frac{\partial^2}{\partial \alpha_1 \partial \alpha_2} \alpha_3 Q + \sum_{\lambda=1}^{3} \kappa_\lambda (\bar{n}_\lambda + 1) \frac{\partial^2 Q}{\partial \alpha_\lambda \partial \alpha_\lambda^*} \Bigg] \\ & + [\text{Compl. conj.}] \Bigg\}. \end{aligned} \tag{15}$$

This equation is still exact (within the approximations of the effective Hamiltonian) and contains derivatives up to second order only. In the laser theory we obtain a generalized Fokker–Planck equation at this stage,[13] which contains derivatives up to infinite order. This is because the number of laser atoms is in fact finite, whereas the interaction parts of the Hamiltonians are equivalent only for an infinite number of atoms.

Let us now consider the second order derivatives of our Fokker–Planck equation which describes the fluctuations. The last term comes from the heat baths. The first term containing second order derivatives comes from the interaction part of the effective Hamiltonian, and shows that even without the heat baths, fluctuations are present due to the parametric interaction of quantized modes. This is just spontaneous parametric emission.

The procedure which we have followed up to now is quite general and may be applied to most of the processes in nonlinear optics. It is especially well adapted to display the dual consequence of nonlinearity; the stimulated coherent effects show up in the first order derivative (drift) terms of eqn (15), and the corresponding spontaneous incoherent effects show up in its second order derivative (diffusion) terms.

We can solve the Fokker–Planck equation by making some approximations. Firstly we assume that the amount of pumping is weak. The amplitudes of signal and idler will then be small and we can neglect the interaction of these modes with the pump mode. The pump mode amplitude is therefore fixed by the driving force and the losses, and is independent of the signal and idler fields generated. This is the parametric approximation. A theory of the parametric amplifier in this approximation has been given by Wagner and Hellwarth.[14] If we also let the damping constants go to zero (this case was first treated by Louisell, Yariv and Siegmann [15, 12]), we obtain exponential gain for both signal and idler. However, this approximation will hold only for a short time at the beginning of the process.

If we include damping and use the parametric approximation we obtain a combined linear Gaussian process for signal and idler. The solution for Q is then a 4-dimensional Gaussian, centred about zero. If $|F_p|^2 < \kappa_3{}^2 \kappa_1 \kappa_2/\beta^2$ then the Gaussian has real width. This is just the condition of weak pumping. The right-hand side of this inequality gives the threshold value of $|F_p|$ for which the system becomes unstable and the parametric approximation will then break down.

We are now able to calculate all interesting multitime correlation functions. According to the Wiener Chinchin theorem, the spectrum of the signal mode is given by the Fourier transform of the correlation function $\langle b_1^\dagger (t+\tau) b_1 (t) \rangle$. Below threshold the signal spectrum is a superposition of two Lorentzians, with the line centre at $\bar{\omega}_1$. One line is weak and broad (linewidth λ_2), the other line is much more intense and narrow (linewidth λ_1). The half-widths at half-power are

$$\lambda_{1/2} = \frac{\kappa_1+\kappa_2}{2} \mp \sqrt{\frac{(\kappa_1+\kappa_2)^2}{4} + \left(\beta^2 \frac{|F_p|^2}{\kappa_3{}^2} - \kappa_1 \kappa_2\right)} \tag{16}$$

(see Fig. 3).

At threshold, λ_1 is zero and the output power

$$P_1 = 2\hbar \bar{\omega}_1 \kappa_1 \langle b_1^\dagger (t) b_1 (t) \rangle \tag{17}$$

diverges. Of course the linear theory breaks down long before threshold. We can eliminate the coupling constant β from eqn (16) and introduce instead P_1, defined by (17). Not too far below threshold, λ_1 depends on $|F_p|$ mainly through the dependence of P_1 on $|F_p|$. Inserting the threshold value of $|F_p|$ everywhere, but not in P_1, we obtain

$$\lambda_1 = 2 \frac{\hbar\omega_1}{P_1} \frac{(\kappa_1 \kappa_2)^2}{(\kappa_1+\kappa_2)^2} (\bar{n}_1 + \bar{n}_2 + 1). \tag{18}$$

This expression is in complete analogy with the expression for the laser linewidth. Corresponding expressions hold, of course, for the idler spectrum.

Let us now consider two possible Hanbury-Brown–Twiss experiments. In the first experiment we absorb the idler frequency in a filter and measure the intensity correlation function of the signal mode, i.e. the quantity

$$K_{11}(\tau) = \frac{\langle b_1^\dagger(t)\, b_1^\dagger(t+\tau)\, b_1(t+\tau)\, b_1(t)\rangle}{|\langle b_1^\dagger(t)\, b_1(t)\rangle|^2}. \tag{19}$$

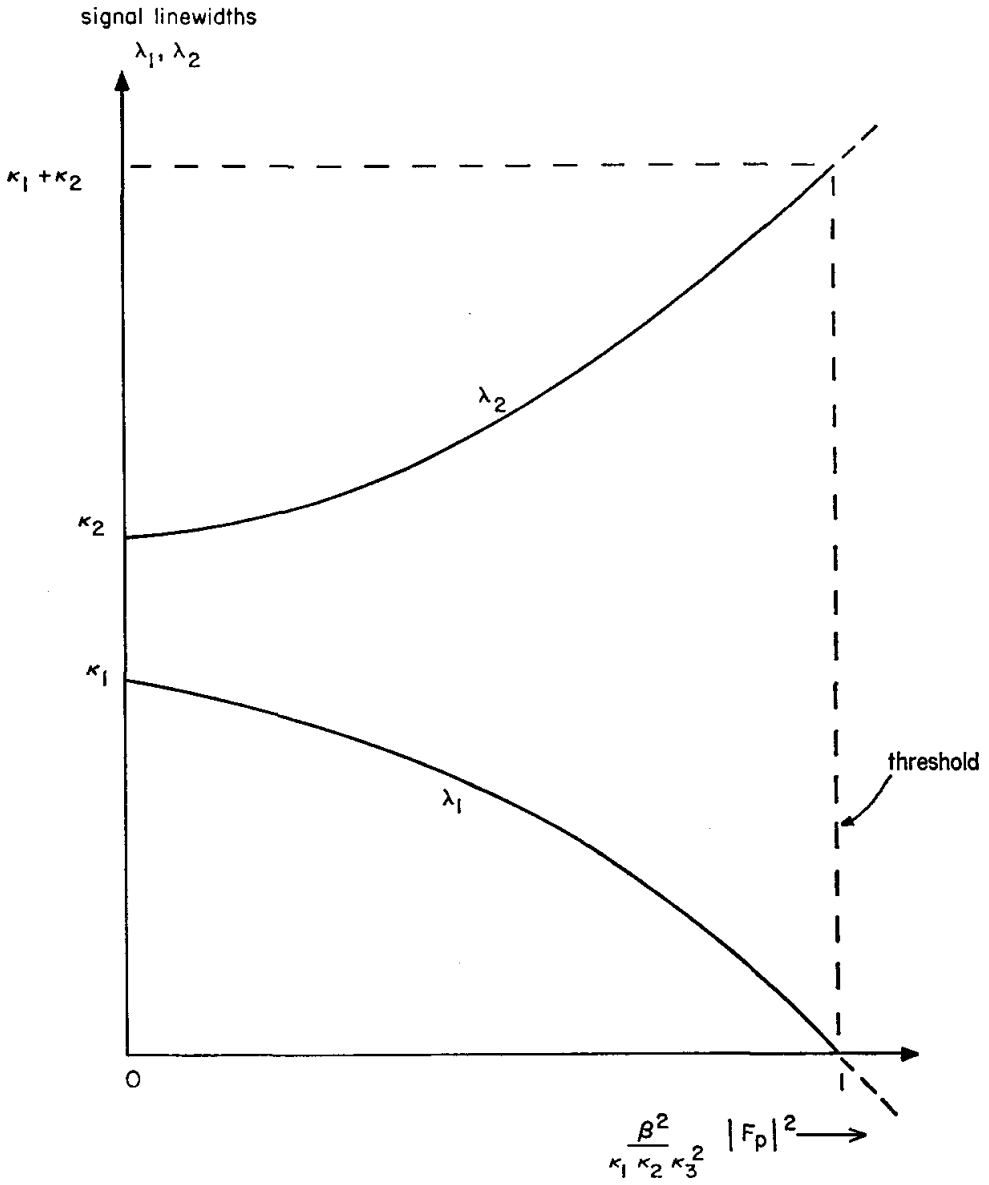


Fig. 3. Linewidths λ_1, λ_2, below threshold, as a function of the pumping force intensity (obtained in the linear theory) shown here for $\kappa_2 > \kappa_1$.

In the second experiment we measure the intensity of the signal mode in one detector and the intensity of the idler mode in a second detector, and correlate them. Thus we would measure the quantity

$$K_{12}(\tau) = \frac{\langle b_1^\dagger(t)\, b_2^\dagger(t+\tau)\, b_2(t+\tau)\, b_1(t)\rangle}{\langle b_1^\dagger(t)\, b_1(t)\rangle \langle b_2^\dagger(t)\, b_2(t)\rangle} \tag{20}$$

We may calculate $K_{11}(\tau)$ and $K_{12}(\tau)$ from the solution of the Fokker–Planck equation. However, as the general expressions are rather lengthy we shall discuss here only the following two limits.

(*a*) Not too far below threshold, only the intense and narrow line of the spectrum will contribute and we obtain

$$K_{11}(\tau) = K_{12}(\tau) = 1+\exp(-2\lambda_1 |\tau|). \tag{21}$$

This shows that in this region of operation, we should observe a Hanbury-Brown–Twiss effect for signal and idler in their autocorrelation functions and in their crosscorrelation function. The width of the correlation function approaches zero when the pumping force F_p approaches the threshold value. If we let the pumping force F_p tend to zero, we obtain (if $\kappa_1 \neq \kappa_2$)

$$K_{11}(\tau) = 1+\exp(-2\lambda_1 |\tau|) \tag{22}$$

$$K_{12}(\tau) = 1+\tfrac{1}{2}\frac{\beta^2 |F_p|^2}{\kappa_3{}^2 (\kappa_1-\kappa_2)^2 \bar{n}_1 \bar{n}_2}\left(\frac{\kappa_1-\kappa_2}{2}+\kappa_1 \bar{n}_1 - \kappa_2 \bar{n}_2\right)^2 \times (\exp(-\lambda_1 |\tau|)+\exp(-\lambda_2 |\tau|))^2. \tag{23}$$

We still observe a Hanbury-Brown–Twiss effect in the autocorrelation function, whereas it disappears in the crosscorrelation function, since signal and idler will be statistically independent (weak) beams in this limit.

(*b*) We now consider the much more interesting case well above threshold, when depletion of the pump cannot be neglected. It is easy to show[7] that a steady state is reached, in which the pump mode is completely saturated and depleted to its threshold value (i.e. independent of F_p) whereas the intensities of signal and idler are determined by F_p.

$$\langle b_1^\dagger b_1\rangle = \sqrt{\frac{\kappa_2}{\kappa_1}}\left(\frac{|F_p|}{\beta} - \frac{\kappa_3 \sqrt{\kappa_1 \kappa_2}}{\beta^2}\right). \tag{24}$$

Their relative intensity is determined by the Manley Rowe relation[2]

$$\frac{P_1}{\omega_1} = \frac{P_2}{\omega_2}. \tag{25}$$

The statistical properties of the parametric oscillator are contained in small phase and amplitude fluctuations about this steady state. Thus, we quasi-linearize the problem with respect to these small fluctuations and obtain a solution of the Fokker–Planck equation for the resulting linear Gaussian process. The spectral linewidth of the amplitude correlation function of the signal is found to be half that given by expression (18). This linewidth is due to slow undamped stochastic motion of the phase difference between signal and idler, arising from spontaneous parametric emission. The sum of signal and idler phases is stable and locked to the phase of the pump.

We are again interested in the theoretical result of the two Hanbury-Brown–Twiss experiments proposed above. As the general expressions for the correlation functions are again rather complicated, we give them only in the region not too far above threshold, i.e. in the limit

$$\langle b_1^\dagger b_1 \rangle \ll \frac{\kappa_2 \kappa_3}{4\beta^2}.$$

Since in general, $\kappa_2 \kappa_3/4\beta^2$ is a very large number, this limit covers all cases of interest. We obtain

$$K_{11}(\tau) = 1 + \left(\frac{\hbar\bar{\omega}_1}{P_1}\right)^2 \frac{2\kappa_1^2 \kappa_2^2 \kappa_3 (\bar{n}_1 + \bar{n}_2 + 1)}{\beta^2 (\kappa_1 + \kappa_2)} \exp(-\lambda|\tau|), \tag{26}$$

$$K_{12}(\tau) = 1 + \left(\frac{\hbar\bar{\omega}_1}{P_1}\right)\left(\frac{\hbar\bar{\omega}_2}{P_2}\right) \frac{2\kappa_1^2 \kappa_2^2 \kappa_3 (\bar{n}_1 + \bar{n}_2 + 1)}{\beta^2 (\kappa_1 + \kappa_2)} \exp(-\lambda|\tau|) \tag{27}$$

with linewidth

$$\lambda = \frac{P_1}{\hbar\bar{\omega}_1} \frac{2\beta^2}{\kappa_3(\kappa_1 + \kappa_2)}. \tag{28}$$

This result shows firstly, that signal and idler both have the same statistical properties as a laser mode. For both modes, the Hanbury-Brown–Twiss effect dies out if we go sufficiently far above threshold. The intensity correlation function then factorizes. Secondly, eqn (27) demonstrates that the Hanbury-Brown–Twiss effect also disappears for the crosscorrelation function sufficiently far above threshold. In fact, besides a trivial normalization factor, $K_{12}(\tau)$ equals $K_{11}(\tau)$. This shows that photons in the signal and idler modes interfere with each other as if they were photons of one coherent mode.

We can make a qualitative comparison between formulae (23) and (27). Both demonstrate the disappearance of the Hanbury-Brown–Twiss effect in the crosscorrelation function, but the reasons for the factorization of the crosscorrelation functions are exactly opposite in the two cases; in (23) the reason is complete incoherence, whilst in (27) it is complete coherence.

Finally, I want to mention that it is also possible to obtain a solution in the threshold region,[16] by symmetrically applying an adiabatic approximation to signal and idler, and eliminating the idler and pump amplitudes. This procedure leads directly to the Fokker–Planck equation for the van der Pol oscillator in the rotating wave approximation, which one also encounters in the laser theory. Therefore solutions for the photon distribution of the signal mode, the transient distribution etc. may be written down immediately. However, it has not been possible so far to derive an analytical theory for the threshold region, which also contains information about correlations between signal and idler. Some further work in this region seems to be necessary, especially when experiments on the photon statistics of parametric oscillators will be done some day in the future.

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Radiation from a System of N Two Level Atoms

D. F. WALLS

There exists a close formal analogy between radiation from a system of N two level atoms[1-4] and the parametric amplification or down-conversion process of nonlinear optics.[5-7] Parametric amplification in a nonlinear dielectric may be described by the following Hamiltonian,

$$H = H_0 + H_1, \tag{1}$$

where

$$\begin{aligned} H_0 &= \hbar\omega_a a^\dagger a + \hbar\omega_b b^\dagger b + \hbar\omega_c c^\dagger c \\ H_1 &= \hbar\kappa\,(a\,b^\dagger c^\dagger + a^\dagger bc) \end{aligned} \tag{2}$$

(a, b, c are the boson annihilation operators for the pump, signal and idler modes respectively. κ is the coupling constant and $\omega_c = \omega_a - \omega_b$). The interaction of N two level atoms with one (resonant) mode of the radiation field may be described by the Hamiltonian (1) with,

$$\begin{aligned} H_0 &= \hbar\omega\, c^\dagger c + \hbar\omega\, J_z \\ H_1 &= \hbar\kappa\,(cJ_+ + c^\dagger J_-) \end{aligned} \tag{3}$$

$$(J_z = \textstyle\sum^N \sigma_z^i,\ J_+ = \sum^N \sigma_i^+,\ J_- = \sum^N \sigma_i^-,$$

where σ_z are the z components of spin, and σ^+, σ^- the spin flip operators for the individual atoms). The angular momentum operators J_z, J_+, and J_- may be represented in terms of two operators a, b obeying boson commutation relations.[8]

$$\begin{aligned} J_z &= \tfrac{1}{2}\,(a^\dagger a - b^\dagger b) \\ J_+ &= ba^\dagger \\ J_- &= b^\dagger a \end{aligned} \tag{4}$$

With this representation the Hamiltonians (2) and (3) become formally identical. We may make the identification; laser pump equivalent to upper atomic level, signal photon equivalent to lower atomic level, idler photon equivalent to emitted photon. One notes that the operators a and b differ from boson operators in that the eigenvalues of $a^\dagger a$ and $b^\dagger b$ assume integer

values from 0 to $2|\mathbf{J}|$. If the total angular momentum $\mathbf{J}$ of the N atom system is a maximum (i.e. $J = N/2$) the eigenvalues of $a^\dagger a$ and $b^\dagger b$ are the actual occupation numbers of the upper and lower levels. This is the case we shall consider. If $J < N/2$ the eigenvalues of $a^\dagger a$ and $b^\dagger b$ are the effective occupation numbers. Only in the case of a large ensemble of atoms, $N \to \infty$, may the operators a and b be identified as true boson operators.

The Heisenberg equations of motion resulting from the Hamiltonian (2) or (3) are nonlinear operator equations. Rather than attempting a direct solution of these equations, we shall search for constants of the motion and solve the Schrödinger equation for the system. Under conditions of perfect energy conservation, $\omega_a = \omega_b + \omega_c$, it can be shown that the Hamiltonians $H_0(t)$ and $H_1(t)$ commute with each other

$$[H_0(t), H_1(t)] = [H(t), H_0(t)] = [H(t), H_1(t)] = 0 \tag{5}$$

Thus a representation exists in which the total Hamiltonian $H(t)$ is diagonal. Once the Hamiltonian has been diagonalized, the Schrödinger equation has been solved.

The eigenstates of H_0 are the number states of the form $|n_a, n_b, n_c\rangle$. If we assume the initial state is $|n_a, n_b, 0\rangle$, the possible eigenstates of H_0 are

$$\Psi = (|n_a, n_b, 0\rangle, |n_a - 1, n_b + 1, 1\rangle \ldots |0, n_a + n_b, n_b\rangle)^+. \tag{6}$$

We shall attempt to express the eigenstates of H_1 as linear combinations of the eigenstates of H_0. The result of H_1 operating on Ψ is

$$H_1 \Psi = \hbar \kappa \tilde{A}_{n_a} \Psi, \tag{7}$$

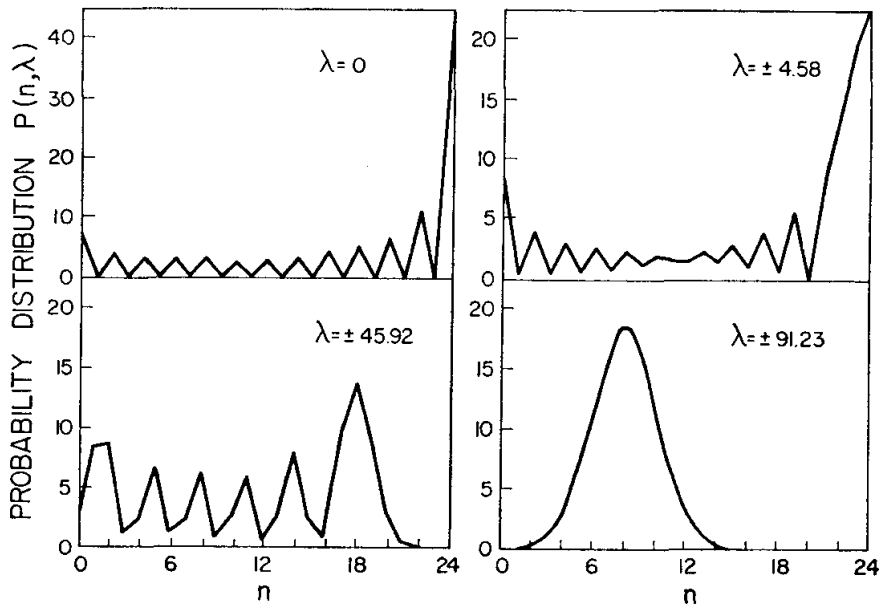


FIG. 1. Plots of $p(n,\lambda)$ against n for $n_a = 24$.

where $\tilde{A}_{n_a}$ is the $(n_a+1)\times(n_a+1)$ symmetric matrix

$$\tilde{A}_{n_a} = \begin{pmatrix} 0 & a_1 & & & & \\ a_1 & 0 & \cdot & & & \\ & \cdot & \cdot & a_r & & \\ & & a_r & 0 & \cdot & \\ & & & \cdot & \cdot & a_{n_a} \\ & & & & a_{n_a} & 0 \end{pmatrix} \tag{8}$$

and $a_r = [(n_a-r+1)\,(n_b+r)]^{\frac{1}{2}}$, $r = 1, 2 \ldots n_a$.

This matrix may be diagonalized numerically and the eigenstates and eigenvalues of the system found. This enables one to calculate the mean number and the probability distribution of the emitted photons. This procedure is described in more detail in Walls and Barakat.[9]

We shall consider first the results for the case of spontaneous emission $M = J$ $(n_b = n_c = 0)$. In Fig. 1 several examples $p\,(n, \lambda)$, the probability that the eigenstate having eigenvalue λ contains n emitted photons, are plotted for $M = J$ $(n_b = n_c = 0)$. In Fig. 1 several examples $p\,(n, \lambda)$, probability that the eigenstate having eigenvalue λ contains n emitted photons, are plotted for $n_a = 24$. The eigenstates with $\lambda = \pm\lambda_{\max}$, that is, the ground state and most highly excited state, have approximately a Poisson distribution in photon

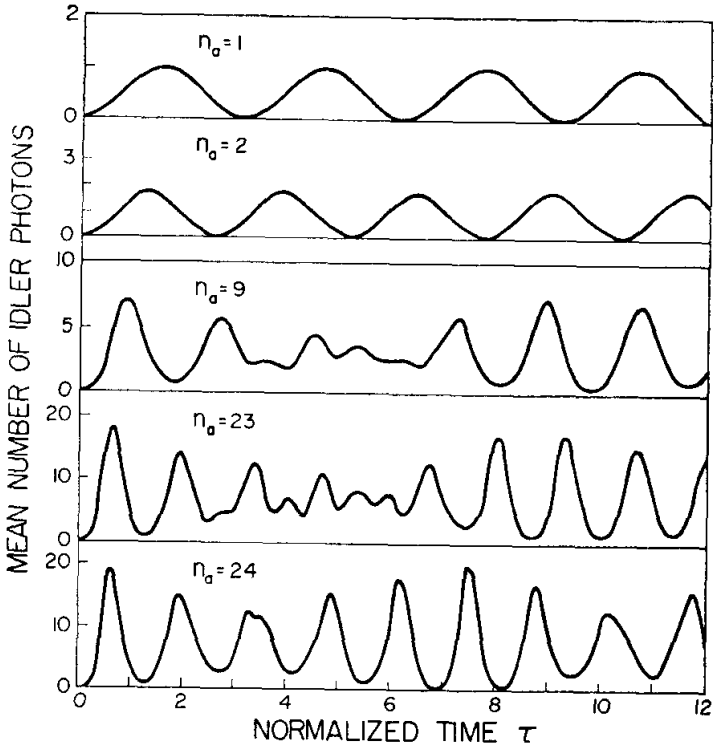


FIG. 2. Mean number of emitted photons as a function of τ, for various values of n_a.

number. The mean number of emitted photons is plotted as a function of $\tau = \kappa t$ in Fig. 2 for $n_a = 1, 2, 9, 23, 24$. We observe the oscillatory (almost periodic) behaviour, in direct contrast to the exponentially increasing solution predicted by the parametric approximation.[6, 7] The fluctuations which appear are due to the amplification of the vacuum fluctuations which have a large effect for this low number of atoms. For a large number of atoms, the vacuum fluctuations, though initially the source of the field, are quickly masked and the mean number of photons follows an elliptic function.[4]

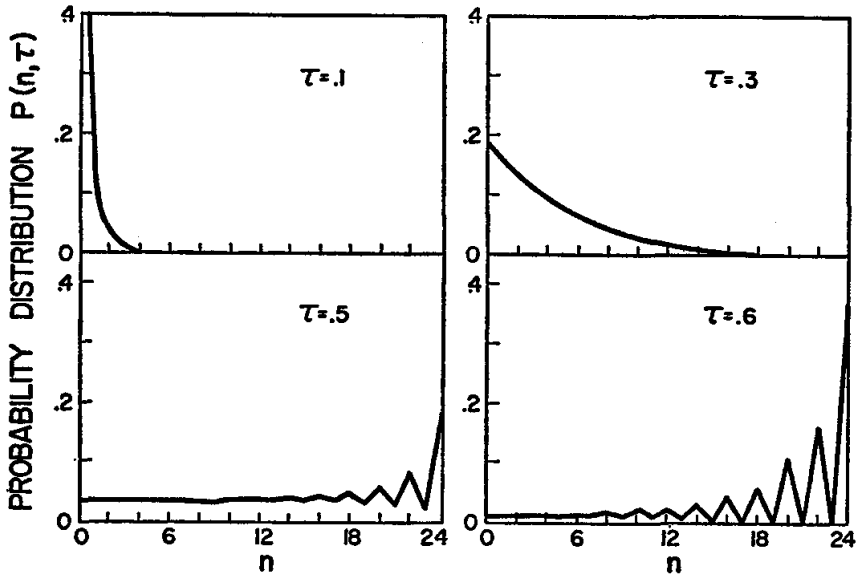


FIG. 3. Probability distribution of emitted photons at several different times, for $n_a = 24$.

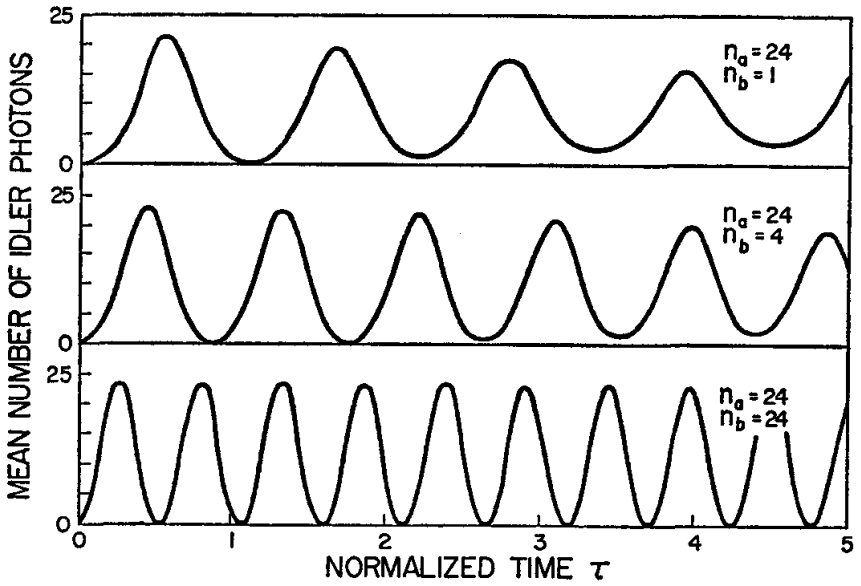


FIG. 4. Mean number of emitted photons in stimulated emission, for $n_a = 24$ and increasing values of n_b.

The probability distribution of the number of emitted photons is plotted in Fig. 3 at several fixed times for $n_a = 24$. For very short times ($\tau \leqslant 0{\cdot}05$) the probability distribution follows a chaotic or power law distribution. However, as the time increases the distribution becomes peaked about increasingly higher values of $\bar{n}_c$, out to the first maximum of $\bar{n}_c$.

In Fig. 4 the behaviour of the mean number of photons for the case of stimulated emission with $n_a = 24$, $n_b = 1, 4, 24$ is shown. We observe a considerable smoothing of the behaviour of $\bar{n}_c(\tau)$ as n_b is increased. For $n_b = 4$, the mean number of idler photons is already very nearly periodic. Thus a finite number of atoms in the ground state serves to mask the effect of the vacuum fluctuations.

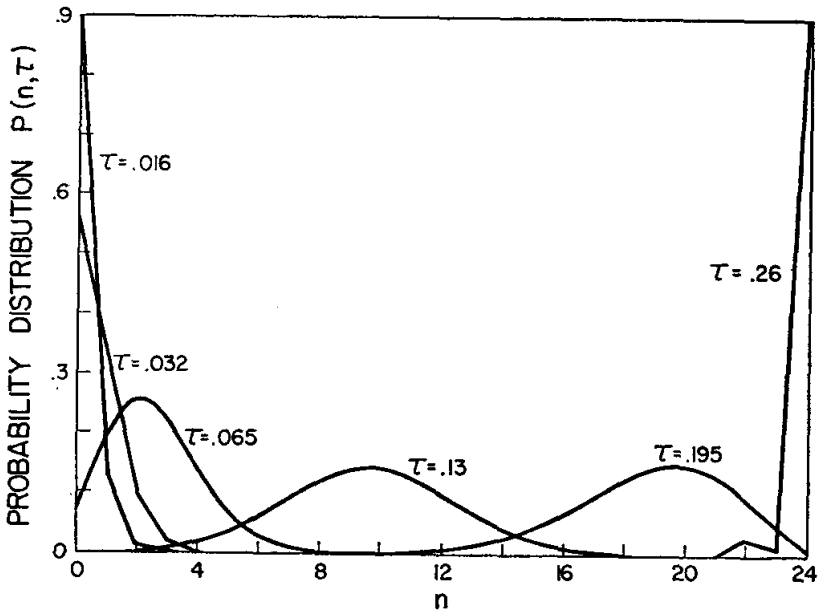


FIG. 5. Probability distributions of emitted photons for the super-radiant case, plotted at several different times.

For $n_a = n_b$, (i.e. $M = 0$) the super-radiant state, $n_c(\tau)$ follows a periodic classical type behaviour. The probability distributions of the emitted photons for the super-radiant case are shown in Fig. 5. For very short times ($\tau \lesssim 0{\cdot}05$) the probability distribution follows a Poisson distribution. This result is expected after observing that in this time range the source of the radiation field (i.e. the transverse component of the total angular momentum J) is essentially classical.

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Non-Adiabatic Effects in the Laser[1]

F. HAAKE

As has been pointed out by Haken,[2] laser theory at the most fundamental level has to deal with a system having a tremendously large number of degrees of freedom; the active atoms interact with the electromagnetic field, and loss and pump mechanisms must be coupled to this "proper" laser system. There is no hope of obtaining an exact solution of the corresponding dynamical problem. It has been an important first step in all laser theories to make an approximation to eliminate the coordinates of the loss and pump mechanisms. The Markoff approximation has turned out to be completely sufficient for this purpose. The "proper" laser system is then described in such a way that it undergoes a quantum mechanical Markoff process. This can be done either by writing down the set of Langevin equations for atomic and field operators[2] or, equivalently, the master equation for the atom–field density operator, or, in the framework of semiclassical theory, a Fokker–Planck equation for a distribution function of the atom and field variables.

It should be noted that all three types of equation resulting from this first step in laser theory still contain more information than is usually needed to explain the experimental results. These results are described in terms of the electromagnetic field alone and never in terms of the active atoms. It is therefore tempting to eliminate the atomic variables too. This has been done many times using the "adiabatic" approximation, (this assumes that the atoms follow the motion of the field adiabatically).

Let us consider two equations arising from such a procedure. Firstly, the well-known van der Pol equation

$$\dot{b}^+ + \{\kappa - \alpha_l + \alpha_{nl}\, b^+ b\}\, b^+ = F^+(t)$$

with

$$\langle F(t)\rangle = 0,\ \langle F^+(t+\tau)\, F(t)\rangle = 4q\,\delta(\tau), \tag{1}$$

and secondly the Fokker–Planck equation

$$\dot{P}(\beta, \beta^*, t) = \left\{\left(\frac{\partial}{\partial\beta}\beta + \frac{\partial}{\partial\beta^*}\beta^*\right)(\kappa - \alpha_l + \alpha_{nl}\,\beta^*\beta) + 4q\,\frac{\partial^2}{\partial\beta^*\partial\beta}\right\} P(\beta, \beta^*, t). \tag{2}$$

In both equations we find terms referring to the damping of the field (κ), the linear gain (α_l), the nonlinearity (α_{nl}), and the fluctuations (q). We also see that the electromagnetic field here is assumed to have Markoffian properties. In eqn (1), this shows up in the δ-function correlation of the fluctuating driving force $F^+(t)$. On the other hand, this property is inherent in the Fokker–Planck equation (2). It is well known that the equations are equivalent. Equation (1) describes the quantum mechanical Markoff process in the Heisenberg picture and eqn (2) describes the same process in the Schrödinger picture.

I will now put forward three objections to the adiabatic or Markoff approximation which underlies these two equations.

1. As mentioned above, the "proper" laser system (active atoms + field) can certainly be treated as undergoing a Markoff process; but the field alone, as a subsystem, cannot. This is a familiar argument in the theory of stochastic processes.

2. Intuitively, we may reason that the active atoms produce an indirect interaction between the photons of the lasing field mode. The situation is then similar to that in superconductivity, where indirect interaction between electrons is produced by phonons. The latter is well known to be a retarded interaction. The retardation effect is neglected in the BCS theory, but has to be retained when more sophisticated properties of super-conducting systems are studied. Correspondingly, an exact elimination of atomic variables in the laser theory must lead to a retarded photon interaction.

3. The problem of eliminating the atomic variables from the "proper" laser equation is somewhat analogous to the problem of eliminating heat bath coordinates when studying the interaction of a microscopic system (harmonic oscillator or atom) with a macroscopic heat bath. The essential condition for elimination of the heat baths in the Markoff approximation is that the heat bath should have a white or at least an extremely broad band energy spectrum. This is just the opposite of the condition sought after in a laser.

These three objections do not help us to decide quantitatively if, and under what conditions, the adiabatic approximation is invalid. To do this, it is necessary to adopt a more sophisticated elimination method. Of course, it is possible to avoid any elimination at all and try to solve for example, the full set of laser Langevin equations. This procedure has been discussed in the chapter by Haken. However, here we use an elimination procedure which is based on Zwanzig's projector formalism.[3]

Let a system be described by a density operator $W(t)$ and consider the decomposition

$$W(t) = pW(t) + (1-p)\,W(t). \tag{3}$$

The algebraic quantity p occurring here may be defined as a projector acting upon $W(t)$ such that:

$$p^2 = p \quad \text{or} \quad (1-p)\,p = 0 \tag{4}$$

Further, let $W(t)$ obey an equation of motion of the form

$$\dot{W}(t) = -i\mathscr{L}\,W(t), \tag{5}$$

where $\mathscr{L}$ is the Liouville operator, chosen according to the physical problem under consideration. Inserting the decomposition (3) into the right-hand side of (5) and then acting from the left, alternatively with p and $(1-p)$, we obtain a system of coupled equations of motion for the two quantities $pW(t)$ and $(1-p)\,W(t)$ respectively. It is an easy matter to eliminate one quantity, say $(1-p)\,W(t)$, from this set. Then the following inhomogeneous integrodifferential equation for the other quantity, $pW(t)$, is obtained.

$$p\dot{W}(t) = -ip\mathscr{L}pW(t) + \int_0^t \mathrm{d}s\,K(s)\,pW(t-s) + \mathscr{I}(t). \tag{6}$$

The inhomogeneity, $\mathscr{I}(t)$, and the kernel, $K(s)$, are given by

$$\begin{aligned} \mathscr{I}(t) &= -ip\mathscr{L}\exp\left[-i(1-p)\mathscr{L}t\right] \times (1-p)\,W(0) \\ K(s) &= -p\mathscr{L}\exp\left[-i(1-p)\mathscr{L}s\right] \times (1-p)\mathscr{L}. \end{aligned} \tag{7}$$

This formalism was first used by Zwanzig himself. He defined the projector, p, such that $pW(t)$ is the diagonal part of the density operator $W(t)$ with respect to some specific representation. However, for the present purpose we follow the method of Argyres and Kelley[4] in which p is defined as the trace over atomic variables of the atom-field density operator for the laser:

$$\begin{aligned} p &= A\,\mathrm{tr}_A \quad \text{with} \quad \mathrm{tr}_A\,A = 1 \\ p\,W(t) &= A\,\mathrm{tr}_A\,W(t) \equiv A\rho(t). \end{aligned} \tag{8}$$

The "parameter" A is an operator in the Hilbert space of the active atoms. It can be chosen arbitrarily within the constraint of normalization which ensures that p is a projector. With this definition of p, eqn (6) can be regarded as an exact equation of motion for the reduced density operator $\rho(t)$ of the laser field. Exact elimination of the atomic variables has thus been achieved. Of course, the Liouvillian $\mathscr{L}$ has to be specified according to the laser problem. Taking eqn (5) as the well-known laser master equation, $\mathscr{L}$ becomes

$$\mathscr{L} = \mathscr{L}_A + \mathscr{L}_F + \mathscr{L}_{AF} + i\Lambda_A + i\Lambda_F. \tag{9}$$

Here $\mathscr{L}_A$, $\mathscr{L}_F$, $\mathscr{L}_{AF}$ are the commutator operations with the Hamiltonians, and refer to the free motion of the active atoms, the field, and the atom–field interaction, respectively. Λ_A and Λ_F represent the incoherent part of the motion of atoms and field according to pump and loss mechanisms (see Haken[2] for an explicit description of these terms).

The equation of motion for the field density operator, eqn (8), cannot be solved exactly. We therefore expand the kernel and the inhomogeneity as a power series, in terms of the interaction Liouvillian $\mathscr{L}_{AF}$, and truncate these series at some finite order. The expansion must be continued up to terms in fourth order of the atom–field interaction ($\mathscr{L}^4{}_{AF}$), since this is the term which contains the nonlinearity necessary to describe laser action. If the expansion were continued only up to terms in $\mathscr{L}^2{}_{AF}$, this would be physically equivalent to considering only the action of the active atoms on the field and neglecting any reaction in the reverse direction. Since saturation effects in the atomic system are most important in the laser, this would be unrealistic.

When expansion is carried out to the correct order, it is found that the time behaviour of the kernel is characterised by four different retardation effects;

1. relaxation of the atomic dipole moment produces a retardation of the form $\exp(-\gamma_\perp s)$;
2. relaxation of the atomic inversion contributes a retardation term $\exp(-\gamma_\parallel s)$;
3. the field damping occurs as $\exp(-\kappa s)$;
4. the inhomogeneous broadening of the atomic line appears as

$$\sum_{\mu=1}^{N} g_\mu{}^2 \exp[-i(\nu_\mu - \omega)s], \qquad \sum_{\mu=1}^{N} g_\mu{}^4 \exp[-i(\nu_\mu - \omega)s].$$

This is the most complex situation to be dealt with in laser theory. In order to show the essential features of the present treatment, we consider the special case of a homogeneous line in resonance ($\nu_\mu = \omega$) and assume that the field damping is small compared to the atomic damping constants ($\kappa \ll \gamma_\perp, \gamma_\parallel$, for example, as in a gas laser). The resulting equation of motion for the field density operator, expressed in terms of the P-representation

$$\rho(t) = \int \mathrm{d}^2\beta P(\beta, \beta^*, t) |\beta\rangle \langle\beta|$$

with respect to coherent states $b|\beta\rangle = \beta|\beta\rangle$, then becomes

$$\dot{P}(\beta, \beta^*, t) = \kappa\left\{\frac{\partial}{\partial\beta}\beta + \frac{\partial}{\partial\beta^*}\beta^* + 2n_{\text{thermal}}\frac{\partial^2}{\partial\beta^*\partial\beta}\right\}P(\beta, \beta^*, t)$$

$$+\int_0^t \mathrm{d}s\underbrace{\left[\left(\frac{\partial}{\partial\beta} + g\frac{\partial}{\partial\beta^*}\beta^*\right)(-\alpha_l\phi_1(s) + \alpha_{nl}\phi_2(s)\beta^*\beta) + 4q\phi_1(s)\frac{\partial^2}{\partial\beta^*\partial\beta}\right]}_{K(s)} \times P(\beta, \beta^*, t-s) \quad (10)$$

There is now a remarkable similarity with the Fokker–Planck equation (2). We again find terms describing the field damping, the linear gain, the non-linearity and the fluctuations. However, we see here that the last three effects occur under a time integral and that they appear retarded as required. The retardation functions $\phi_1(s)$, $\phi_2(s)$ are given by

$$\phi_1(s) = \gamma_\perp \exp(-\gamma_\perp s)$$

$$\phi_2(s) = \frac{\gamma_\perp{}^2\gamma_\parallel}{(\gamma_\perp - \gamma_\parallel)^2}[\exp(-\gamma_\parallel s) - \{1 + s(\gamma_\perp - \gamma_\parallel)\}\exp(-\gamma_\perp s)]. \quad (11)$$

In eqn (10), the inhomogeneity has been omitted. $\mathscr{F}(t)$ turns out to decay as $\exp(-\gamma_\perp t)$ and therefore does not play any role in the laser operating under stationary conditions. It is however, important for a discussion of transient effects and we see by inspection of eqn (7) that it depends on the initial conditions in the laser. Some correction terms to the kernel $K(s)$, in particular those which contain field variable derivatives up to fourth order, have also been neglected. We can see that this is valid if we normalize the field variables, viz;

$$\bar{\beta} = \left(\frac{\alpha_{nl}}{q}\right)^{\frac{1}{2}}\beta, \qquad \bar{\beta}^* = \left(\frac{\alpha_{nl}}{q}\right)^{\frac{1}{2}}\beta^*,$$

The correction terms then contain

$$\sqrt{\frac{\alpha_{nl}}{q}} \approx [\text{photon number at threshold}]^{-1} \ll 1.$$

We now turn to the solution of the integrodifferential equation (10). The kernel $K(s)$ in this equation obeys the following differential equation, as a function of time

$$\dddot{K}(s)+(2\gamma_\perp+\gamma_\parallel)\,\ddot{K}(s)+\gamma_\perp\,(\gamma_\perp+2\gamma_\parallel)\,\dot{K}(s)+\gamma_\perp{}^2\,\gamma_\parallel\,K(s)=0. \qquad (12)$$

Differentiating eqn (10) with respect to t and linearly combining the resulting equations for $\dot{P}(t)$, $\ddot{P}(t)$, $\dddot{P}(t)$ and $\ddddot{P}(t)$ according to eqn (12), we find that the P-function obeys a fourth order differential equation in time. A simplified version of this equation becomes

$$\dddot{P}(t)+(2\gamma_\perp+\gamma_\parallel)\,\ddot{P}(t)+\gamma_\perp\,(\gamma_\perp+2\gamma_\parallel)\,\ddot{P}(t)+\gamma_\perp{}^2\,\gamma_\parallel\,\dot{P}(t)=\gamma_\perp{}^2\,\gamma_\parallel\,\Lambda P(t), \quad (13)$$

where Λ is an abbreviation for the Fokker–Planck differential operator occurring on the right-hand side of eqn (2).

Now we may discuss the non-adiabatic or non-Markoffian effects of the laser field mode. The eigenvalues and eigenfunctions of Λ are known from Risken's work

$$\Lambda f_{nm}(\beta,\beta^*)=\gamma_{nm}\,f_{nm}(\beta,\beta^*); \qquad n=0,\pm1,\pm2,\ldots \quad m=0,1,2,\ldots \quad (14)$$

We are left with the problem of finding the time dependence of the general solution

$$P(\beta,\beta^*,t)=\sum_{n=-\infty}^{+\infty}\sum_{m=0}^{\infty}A_{nm}T_{nm}(t)f_{nm}(\beta,\beta^*)$$

The secular equation for $T_{nm}(t)$ has four roots which depend on the relative sizes of $\gamma_\perp$, $\gamma_\parallel$, γ_{nm}. Near threshold, $\gamma_{nm}\ll\gamma_\perp,\gamma_\parallel$ for all experimentally interesting γ_{nm}. One root then practically coincides with γ_{nm}, the others being very much smaller. In this case the second and higher order time derivatives in (10) are completely negligible and (10) reduces to the Fokker–Planck equation (2). On the other hand, the four roots become comparable in magnitude as γ_{nm} approaches $\gamma_\perp,\gamma_\parallel$. For γ_{nm} close to $\gamma_\perp,\gamma_\parallel$ the roots become complex. Results of Risken and Vollmer show that the latter condition can indeed be fulfilled sufficiently far above threshold for some γ_{nm}. As a result, some observables (for example, the intensity correlation function) will no longer approach their equilibrium values monotonically but will show damped oscillations. An experimental investigation of these effects should be very interesting.

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Determination of the Statistical Properties of Light from Photoelectric Measurements

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1. INTRODUCTION

It is well known that photoelectric measurements yield information about the statistical properties of fluctuating light beams. Arecchi, Pike and Mandel and their co-workers, together with many others, have done a lot of work in this field.[1-13] Such measurements give us the distribution of photoelectrons emitted within a fixed time interval T, from a photodetector upon which the light beam is incident normally. This distribution is identical, as we shall show later to the distribution of absorbed photons and it can therefore be called a photon-counting distribution. Alternatively, the measurements provide the moments of this photon-counting distribution, from which the factorial moments can easily be derived using the following relation, (2).

If $\langle W^k \rangle_{\mathcal{N}}$ is the moment of the probability distribution $P_{\mathcal{N}}(W)$ of the time integrated intensity W, given by

$$W = \int_0^T I(t')\,\mathrm{d}t', \tag{1}$$

then

$$\langle W^k \rangle_{\mathcal{N}} = \left\langle \frac{n!}{(n-k)!} \right\rangle = \sum_{n=0}^{\infty} p(n)\, n(n-1)\ldots(n-k+1)$$

$$= a_1^{(k)} \langle n \rangle + \ldots + a_k^{(k)} \langle n^k \rangle, \tag{2}$$

where $p(n)$ is the photon-counting distribution, $\langle n^k \rangle$ are its moments, $\langle n!/(n-k)! \rangle$ are its factorial moments and

$$a_j^{(k)} = (-1)^{k-j} \sum (x_1 x_2 \ldots x_{k-j}), \qquad j = 1, 2, \ldots, k-1, \quad a_k^{(k)} = 1. \tag{3}$$

The sum is taken over all choices of the numbers $x_1, x_2, \ldots, x_{k-j}$ from the sequence $1, 2, \ldots, k-1$ regardless of the order.

However, in order to understand the statistical behaviour of light, especially laser light, we want to obtain information about the distribution

$P_{\mathscr{N}}(W)$ of the integrated intensity from the photon-counting distribution $p(n)$ itself or its factorial moments. If the time interval T of observation is much smaller than the coherence time, $P_{\mathscr{N}}(W)$ will give us the distribution $P_{\mathscr{N}}(I)$ of the intensity I. The main purpose of this paper is to solve this problem, which from a mathematical point of view, means inverting the photodetection equation

$$p(n) = \int_0^\infty P_{\mathscr{N}}(W) \frac{(\alpha W)^n}{n!} \exp(-\alpha W)\, \mathrm{d}W. \tag{4}$$

Here, the photon-counting distribution $p(n)$ is expressed in terms of the integrated intensity distribution $P_{\mathscr{N}}(W)$, and α is the photoefficiency of the detector. This equation was derived by Mandel[14] on the basis of a semiclassical theory, and by Glauber[15] on the basis of a quantum theory.

We shall show first of all that the distribution of photons absorbed by a photodetector is of the same form as the distribution of emitted photoelectrons. This is a consequence of the definition of the number operator in the theory of the quantized electromagnetic field. We shall also show how the integrated intensity distribution can be determined from the photon-counting distribution or its factorial moments, using a generating function and orthogonal (Laguerre) polynomials. We will then give conditions which must be placed on the factorial moments of the photon-counting distribution (or on the photon-counting distribution itself), so that it is possible to recognize the distribution $P_{\mathscr{N}}(W)$ as either a non-negative ordinary function or a generalized function whose support is composed of a finite number of points.

Using a formalism of arbitrary ordering of field operators, recently introduced into quantum optics by Agarwal and Wolf,[16] and, in particular, the parametrization of this ordering, introduced by Cahill and Glauber,[17] we can derive the general photodetection equation, which expresses $p(n)$ in terms of the distribution $P(W, s)$, related to s-ordering of the field operators (Peřina and Horák[18]).

The methods described will be demonstrated by giving examples of one-mode and also multi-mode superposition of coherent and thermal fields (Peřina and Mista,[19] Peřina and Horák[20]). In particular, we derive the quantities $\langle W^k \rangle_{\mathscr{N}}$, $p(n)$, and $P_{\mathscr{N}}(W)$, taking arbitrary mean occupation numbers per mode. As a special case we consider these quantities when the mean frequencies of the thermal and coherent fields are different. This situation arises in heterodyne detection of thermal light. Results obtained for $s = -1$ (antinormal ordering) could be useful for detection of the field by means of so-called "quantum counters," which operate by stimulated emission rather than by absorption.

2. The Photodetection Equation

In order to show that the photodetection equation arises out of the properties of the quantized electromagnetic field, we define the number operator

$$\hat{n} = \sum_{j=1}^{3} \int_{L^3} \hat{A}_j^{\dagger}(x)\, \hat{A}_j(x)\, \mathrm{d}^3\mathbf{x}. \tag{5}$$

The detection operator, $\hat{A}(x)$ (closely connected with the operators of the electromagnetic field strength) is given by[21,22]

$$\hat{A}_j(x) = L^{-3/2} \sum_{\mathbf{k}} \sum_{s=1}^{2} e_j^{(s)}(\mathbf{k})\, \hat{a}_{\mathbf{k}s}\, \exp\left[i(\mathbf{k}.\mathbf{x} - ckt)\right]. \tag{6}$$

Here, $\hat{a}_{\mathbf{k}s}$ is the annihilation operator of a photon in the wave-vector spin mode $(\mathbf{k}, s) \equiv \lambda$; $e^{(s)}(\mathbf{k})$ is the unit polarization vector. We consider here the number operator defined over a normalization volume L^3. We could equally well define this operator over an arbitrary volume V. All the results given below would still be valid (Mandel,[23] Peřina[24]).

Using (6) we can rewrite (5) in the form;

$$\hat{n} = \sum_{\lambda} \hat{a}_{\lambda}^{\dagger}\, \hat{a}_{\lambda}. \tag{7}$$

We can also introduce the expectation value of $\hat{n}$ in the coherent state $|\{\alpha_\lambda\}\rangle$

$$W = \sum_j \int_{L^3} \mathscr{A}_j^*(x)\, \mathscr{A}_j(x)\, \mathrm{d}^3\mathbf{x} = \sum_{\lambda} |\alpha_\lambda|^2, \tag{8}$$

where

$$\mathscr{A}_j(x) = L^{-3/2} \sum_{\mathbf{k}} \sum_{s=1}^{2} e_j^{(s)}(\mathbf{k})\, \alpha_{\mathbf{k}s} \exp\left[i(\mathbf{k}.\mathbf{x} - ckt)\right] \tag{9}$$

and the complex number $\alpha_{\mathbf{k}s}$ is given by

$$\hat{a}_{\mathbf{k}s} |\{\alpha_{\mathbf{k}s}\}\rangle = \alpha_{\mathbf{k}s} |\{\alpha_{\mathbf{k}s}\}\rangle. \tag{10}$$

We express the density matrix in terms of the Fock and coherent states[25,21,26]

$$\hat{\rho} = \sum_{n,m} \rho(\{n_\lambda\}, \{m_\lambda\})\, |\{n_\lambda\}\rangle\langle\{m_\lambda\}| = \int \phi_{\mathscr{N}}(\{\alpha_\lambda\})\, |\{\alpha_\lambda\}\rangle\langle\{\alpha_\lambda\}|\, \mathrm{d}^2\{\alpha_\lambda\}. \tag{11}$$

$\rho(\{n_\lambda\}, \{m_\lambda\})$ are the Fock matrix elements of the density matrix and $\phi_{\mathscr{N}}(\{\alpha_\lambda\})$ is a weighting function (this is assumed to exist at least as a generalized function for the fields under consideration). Using the commutation rules and

$\rho(\{n_\lambda\}) \equiv \rho(\{n_\lambda\}, \{n_\lambda\})$ we can then write for the moment $\langle \hat{n}^k \rangle$

$$\langle \hat{n}^k \rangle = \sum_{\{n_\lambda\}} \rho(\{n_\lambda\}) \langle \{n_\lambda\} | \hat{n}^k | \{n_\lambda\} \rangle$$

$$= \int \phi_{\mathcal{N}}(\{\alpha_\lambda\}) \langle \{\alpha_\lambda\} | \mathcal{N}_c \left\{ \sum_j \int_{L^3} \hat{A}_j^\dagger(x)\, \hat{A}_j(x)\, \mathrm{d}^3\mathbf{x} \right\}^k | \{\alpha_\lambda\} \rangle \mathrm{d}^2\{\alpha_\lambda\}, \tag{12}$$

where $\mathcal{N}_c$ indicates the operation of normal ordering. Substituting the unit operator $\sum_{\{n_\lambda\}} |\{n_\lambda\}\rangle\langle\{n_\lambda\}| = \hat{1}$ into (12) and using the identity

$$|\langle \{n_\lambda\} | \{\alpha_\lambda\} \rangle|^2 = \prod_\lambda \frac{|\alpha_\lambda|^{2n_\lambda}}{n_\lambda!} \exp(-|\alpha_\lambda|^2) \tag{13}$$

we obtain

$$\sum_{n=0}^{\infty} p(n)\, n^k = \sum_{\{n_\lambda\}} \left\{ \int \phi_{\mathcal{N}}(\{\alpha_\lambda\}) \prod_\lambda \frac{|\alpha_\lambda|^{2n_\lambda}}{n_\lambda!} \exp(-|\alpha_\lambda|^2)\, \mathrm{d}^2\{\alpha_\lambda\} \right\} n^k, \tag{14}$$

where

$$p(n) = \sum_{\sum_\lambda n_\lambda = n} \rho(\{n_\lambda\}) \tag{15}$$

and $\hat{n}|\{n_\lambda\}\rangle = n|\{n_\lambda\}\rangle$.

From (14) it follows that

$$p(n) = \sum_{\sum_\lambda n_\lambda = n} \int \phi_{\mathcal{N}}(\{\alpha_\lambda\}) \prod_\lambda \frac{|\alpha_\lambda|^{2n_\lambda}}{n_\lambda!} \exp(-|\alpha_\lambda|^2)\, \mathrm{d}^2\{\alpha_\lambda\}. \tag{16}$$

Using the multinomial theorem

$$\sum_{\sum_\lambda n_\lambda = n} \prod_\lambda \frac{x^{n_\lambda}}{n_\lambda!} = \frac{(\sum_\lambda x_\lambda)^n}{n!}, \tag{17}$$

we obtain[27,28]

$$p(n) = \int \phi_{\mathcal{N}}(\{\alpha_\lambda\}) \frac{W^n}{n!} \exp(-W)\, \mathrm{d}^2\{\alpha_\lambda\}. \tag{18}$$

Then substituting

$$P_{\mathcal{N}}(W') = \int \phi_{\mathcal{N}}(\{\alpha_\lambda\})\, \delta\left(W' - \sum_\lambda |\alpha_\lambda|^2\right) \mathrm{d}^2\{\alpha_\lambda\}, \tag{19}$$

we finally arrive at

$$p(n) = \int_0^\infty P_{\mathscr{N}}(W) \frac{W^n}{n!} \exp(-W)\, \mathrm{d}W. \tag{20}$$

This is the photodetection equation.

We have thus obtained the same relation for the distribution of the number of photons in the volume L^3, as for the distribution of photoelectrons emitted within the time interval T. Thus, although it is the statistical properties of photoelectrons which are studied in photoelectric measurements, these directly reflect the statistical properties of the photons themselves.

The situation would be completely different if the field were detected by a so-called "quantum counter",[23,24] operating by stimulated emission rather than by absorption. These detectors measure antinormally ordered products of field operators, so that the commutation rules for re-ordering of normally ordered products to antinormally ordered products give the contribution of the physical vacuum through the commutators. (In this case, since

$$\sum_j \int \hat{A}_j^\dagger \hat{A}_j \, \mathrm{d}^3\mathbf{x} = \sum_j \int \hat{A}_j \hat{A}_j^\dagger \, \mathrm{d}^3\mathbf{x} - M,$$

through the number of modes M.) The distribution of counts $p'(n)$ is given by the same relation as (4) with $P_{\mathscr{N}}(W) \to P_{\mathscr{A}}(W)$ in antinormal ordering[23]

$$p'(n) = \int_0^\infty P_{\mathscr{A}}(W) \frac{W^n}{n!} \exp(-W)\, \mathrm{d}W, \tag{21}$$

but in this case, the distribution of photons is given by[24]

$$p(n) = \left(\frac{\mathrm{d}}{\mathrm{d}W}\right)^{n+M-1} [P_{\mathscr{A}}(W) \exp(W)]_{W=0} \tag{22}$$

so that

$$P_{\mathscr{A}}(W) = \exp(-W) \sum_{n=0}^{\infty} \frac{W^{n+M-1}}{(n+M-1)!} p(n). \tag{23}$$

The relation between p' and p is obtained by substituting (23) into (21)[24]

$$p'(n) = \sum_{n'=0}^{\infty} p(n) 2^{-n'-n-M} \binom{n'+n+M-1}{n}. \tag{24}$$

3. Determination of the Integrated Intensity Distribution from the Photon–Counting Distribution

To determine the statistical properties of light from photon-counting statistics, it is necessary to invert the photodetection equation (4) (we shall consider $\alpha = 1$ for simplicity). Denoting $n!p(n)$ by $q(n)$ and $P_{\mathcal{W}}(W)\exp(-W)$ by $Q(W)$, we see that this problem can be reduced to the moment problem, i.e. given the moment sequence

$$\int_0^\infty Q(W)\,W^k\,\mathrm{d}W = \langle W^k\rangle, \qquad k = 0, 1, \ldots \tag{25}$$

we wish to determine the distribution $Q(W)$. Thus, determination of $P_{\mathcal{W}}(W)$ from its moments (i.e. from the factorial moments of $p(n)$) is the same kind of problem as the determination of $P_{\mathcal{W}}(W)$ from $p(n)$ itself. We shall confine ourselves mainly to the determination of $P_{\mathcal{W}}(W)$ from $p(n)$.

We can see that a formal solution of (4) may be written in the form[28]

$$P_{\mathcal{W}}(W) = \exp(W)\sum_{n=0}^{\infty}(-1)^n p(n)\,\delta^{(n)}(W), \tag{26}$$

where $\delta^{(n)}(W)$ is the nth derivative of the Dirac function.

3.1. Method Using an Analytical Generating Function[29]

In general, a mathematical expression such as (26), with an infinite number of terms, may be meaningless. Such a series cannot usually be considered as a generalized function, but it can be considered as a so-called ultradistribution (we will return to this point later). However, here the series does have meaning, because the characteristic generating function is an analytical function,

$$\langle\exp(ixW)\rangle_{\mathcal{W}} = \int_0^\infty P_{\mathcal{W}}(W)\exp(ixW)\,\mathrm{d}W. \tag{27}$$

This is because $P_{\mathcal{W}}(W) = 0$ for $W < 0$. Expressing $\delta^{(n)}(W)$ in (26) as a Fourier integral,

$$\delta^{(n)}(W) = \frac{1}{2\pi}\int_{-\infty}^{\infty}(ix)^n\exp(ixW)\,\mathrm{d}x, \tag{28}$$

we obtain

$$P_{\mathcal{W}}(W) = \frac{1}{2\pi}\exp(W)\int_{-\infty}^{\infty}G(x)\exp(ixW)\,\mathrm{d}x, \tag{29}$$

where

$$G(x) = \sum_{n=0}^{\infty}(-ix)^n p(n). \tag{30}$$

This is the generating function of the distribution $P_{\mathcal{N}}(W)\exp(-W)$. This method was proposed by Wolf and Mehta.[29] The analytical form of the generating function $G(x)$ makes it possible to obtain the values of $G(x)$ for all x by means of an analytical continuation if the series (30) has a finite radius of convergence. Thus, $P_{\mathcal{N}}(W)$ is unique.

For example, for the Bose–Einstein distribution we have

$$p(n) = \langle n\rangle^n/(1+\langle n\rangle)^{1+n},$$

and we get

$$G(x) = (1+\langle n\rangle + ix\langle n\rangle)^{-1} \text{ and } P_{\mathcal{N}}(W) = \langle n\rangle^{-1}\exp(-W/\langle n\rangle).$$

For the Poisson distribution, we have

$$p(n) = \langle n\rangle^n \exp(-\langle n\rangle)/n!$$

and we get

$$G(x) = \exp[-\langle n\rangle(1+ix)] \text{ and } P_{\mathcal{N}}(W) = \delta(W-\langle n\rangle).$$

If the factorial moments of $p(n)$ are given, the moment problem (25) can be solved in an exactly similar way.

3.2. METHOD OF ORTHOGONAL POLYNOMIALS[30–34]

Since every term of (26) represents a generalized function with one-point support, we must know all the $p(n)$ to be able to reconstruct $P_{\mathcal{N}}(W)$ as an ordinary function. This cannot be done in practice. Therefore, we give here a method based on the decomposition of $P_{\mathcal{N}}(W)$ in terms of Laguerre polynomials, in which every term has the whole complex plane as a support. Such a prescription can be used to determine $P_{\mathcal{N}}(W)$ approximately, if a finite number of $p(n)$ (or its factorial moments) are known from experiment.

Firstly, we consider the one-mode case with phase information, so that we invert the formula[21,25]

$$\rho(n,m) = \int \frac{\alpha^n \alpha^{*m}}{\sqrt{(n!m!)}} \exp(-|\alpha|^2)\,\phi_{\mathcal{N}}(\alpha)\,\mathrm{d}^2\alpha, \tag{31}$$

where $\rho(n,m)$ are the Fock matrix elements of the density matrix and $\phi_{\mathcal{N}}(\alpha)$ is the weighting function of the "diagonal" representation of the density matrix in terms of coherent states. This problem was solved by Sudarshan[25] in the form of a double series of the derivatives of the δ-functions. From the mathematical point of view, such a series usually cannot be considered as a generalized function. For this reason, Sudarshan's original prescription for constructing $\phi_{\mathcal{N}}(\alpha)$ was criticized by a number of authors. Furthermore, it was shown by Cahill[35] that such a series does not lead to a distribution on

the D_2-space of testing functions if an infinite number of photons are present in the field. However, it was also shown by Miller and Mishkin[36] that an infinite series of successively higher derivatives of the δ-functions can give rise to a distribution on the Z_2-space of testing functions, which is the Fourier transform of D_2. Mehta and Sudarshan,[37] and Klauder, McKenna and Currie[38] (see also Klauder and Sudarshan[26]) have shown that all density matrices can be expressed as limits of sequences, whose members each have a diagonal representation. These sequences can be composed of a finite number of derivatives of the δ-functions or of square integrable functions. Deeper insight into this problem of the existence of the $\phi_{\mathcal{N}}$-function was given by Cahill and Glauber.[17] They have shown, on the basis of a kind of arbitrary ordering of field operators (s-ordering), the way in which the singularities of the $\phi_{\mathcal{N}}$-function appear. Cahill[39] has proposed some rules for regularization of the $\phi_{\mathcal{N}}$-function.

We note here that we can also obtain some results concerning this problem from our decomposition of $\phi_{\mathcal{N}}$ in terms of Laguerre polynomials. Let us consider the function $\phi_{\mathcal{N}}(\alpha)$ in the form[33]

$$\phi_{\mathcal{N}}(\alpha) = \exp\left[-(\xi-1)|\alpha|^2\right] \sum_{j=0}^{\infty} \left\{ \sum_{\lambda=0}^{\infty} c_{j\lambda} (\xi\alpha)^{\lambda} L_j^{\lambda}(\xi|\alpha|^2) + \sum_{\lambda=1}^{\infty} c_{j\lambda}^{*} (\xi\alpha^{*})^{\lambda} L_j^{\lambda}(\xi|\alpha|^2) \right\}, \tag{32}$$

where $\xi \geqslant 1$ is a real number and L_j^{λ} are the Laguerre polynomials. Multiplying (32) by $\alpha^{*\lambda'} L_k^{\lambda'}(\xi|\alpha|^2) \exp(-|\alpha|^2)$, integrating over $\xi\alpha$ and using the orthogonality condition for the Laguerre polynomials, viz.

$$\int_0^{\infty} x^{\lambda} \exp(-x) L_j^{\lambda}(x) L_k^{\lambda}(x)\, \mathrm{d}x = \delta_{jk} \frac{[\Gamma(j+\lambda+1)]^3}{\Gamma(j+1)}, \tag{33}$$

we obtain the coefficients $c_{j\lambda}$ in terms of the Fock elements $\rho(n, m)$

$$\begin{aligned} c_{j\lambda} &= \frac{j!}{\pi[(j+\lambda)!]^3} \int \phi_{\mathcal{N}}(\alpha) \exp(-|\alpha|^2)\, \alpha^{*\lambda} L_j^{\lambda}(\xi|\alpha|^2)\, \mathrm{d}^2(\xi\alpha) \\ &= \frac{\xi}{\pi(j+\lambda)!} \sum_{s=0}^{j} (-1)^s \binom{j}{s} \left(\frac{s!}{(s+\lambda)!}\right)^{\frac{1}{2}} \xi^s \rho(s, s+\lambda), \end{aligned} \tag{34}$$

where we have used (31) and

$$L_j^{\lambda}(x) = [(j+\lambda)!]^2 \sum_{s=0}^{j} \frac{(-x)^s}{s!(j-s)!\,(s+\lambda)!}. \tag{35}$$

Equation (34) can be inverted into the form

$$\rho(s, s+\lambda) = \frac{\pi}{\xi^{s+1}} \left(\frac{(s+\lambda)!}{s!} \right)^{\frac{1}{2}} \sum_{j=0}^{s} (-1)^j \binom{s}{j} (j+\lambda)!\, c_{j\lambda}\,. \tag{36}$$

We see from (32) that

$$\int \phi_{\mathcal{N}}{}^2(\alpha)\, \mathrm{d}\mu(\alpha) = \pi \sum_{j=0}^{\infty} \sum_{\lambda=0}^{\infty} |c_{j\lambda}|^2 \frac{[(j+\lambda)!]^3}{j!} + \pi \sum_{j=0}^{\infty} \sum_{\lambda=1}^{\infty} |c_{j\lambda}|^2 \frac{[(j+\lambda)!]^3}{j!}, \tag{37}$$

where we have put $\xi = 1$ and $\mathrm{d}\mu(\alpha) = \exp(-|\alpha|^2)\, \mathrm{d}^2\alpha$.

Hence, if (37) is finite, then $\phi_{\mathcal{N}}(\alpha)$ is an L_2-function in the space $\mathrm{d}\mu$, and the series (32) converges to $\phi_{\mathcal{N}}(\alpha)$ in the norm of L_2, i.e.

$$\lim_{N\to\infty} \int (\phi_{\mathcal{N}} - \phi_{\mathcal{N}}{}^{(N)})^2\, \mathrm{d}\mu = 0,$$

where $\phi_{\mathcal{N}}{}^{(N)}$ is a partial sum of (32). In this way, we can determine from (34) a class of physical fields, specified by $\rho(n, m)$, for which $\phi_{\mathcal{N}}(\alpha)$ is a member of the L_2-space.[34] Also, using (32) we can construct sequences, whose members lie in L_2, which converge to the diagonal weighting function $\phi_{\mathcal{N}}(\alpha)$.

For example, for the Gaussian field specified by $\rho(n, m) = \delta_{nm} \cdot \langle n_T \rangle^n / (1 + \langle n_T \rangle)^{1+n}$, for which $\phi_{\mathcal{N}}(\alpha)$ is an ordinary function $((\pi \langle n_T \rangle)^{-1} \exp(-|\alpha|^2/\langle n_T \rangle))$, we have $c_{j0} = (1/j!)(1 + \langle n_T \rangle)^{-j-1}$, and (37) gives

$$\sum_{j=0}^{\infty} [(1 + \langle n_T \rangle)^2]^{-j-1} = (\langle n_T \rangle^2 + 2\langle n_T \rangle)^{-1},$$

which is indeed finite.

We give two examples of states, for which (32) leads to a generalized function.

a. The coherent state $|\beta\rangle$:

$$\phi_{\mathcal{N}}(\alpha) = \delta(\alpha - \beta) = \lim_{M\to\infty} \frac{1}{\pi} \exp(-|\beta|^2) \times \sum_{j=0}^{M} \left\{ \sum_{\lambda=0}^{M} \frac{j!}{[(j+\lambda)!]^3} (\beta\alpha^*)^\lambda L_j^\lambda(|\alpha|^2) L_j^\lambda(|\beta|^2) + \sum_{\lambda=1}^{M} (c.c.) \right\} \tag{38}$$

and if it is averaged over the phase of α:

$$\phi_{\mathcal{N}}(|\alpha|) = \delta(|\alpha|^2 - |\beta|^2) = \lim_{M\to\infty} \frac{1}{\pi} \exp(-|\beta|^2) \sum_{j=0}^{M} \frac{1}{(j!)^2} L_j^0(|\alpha|^2) L_j^0(|\beta|^2). \tag{39}$$

b. The Fock state $|n\rangle$:

$$\phi_{\mathcal{N}}(|\alpha|) = (-1)^n \delta^{(n)}(|\alpha|^2) \exp(-|\alpha|^2) = \lim_{M\to\infty} \frac{1}{\pi} \sum_{j=n}^{M} \frac{(-1)^n}{n!(j-n)!} L_j^0(|\alpha|^2). \tag{40}$$

Since (4) does not depend explicitly on the number of modes M, putting $\lambda = 0$ in (32) we obtain

$$P_{\mathcal{N}}(W) = \exp[-(\xi - 1) W] \sum_{j=0}^{\infty} c_j L_j(\xi W), \tag{41}$$

where $L_j \equiv L_j^0$, and from (34)

$$c_j \equiv c_{j0} = \frac{\xi}{j!} \sum_{s=0}^{j} (-1)^s \binom{j}{s} \xi^s p(s). \tag{42}$$

If only the first N terms of $p(n)$ are known, the accuracy of this approximation is given by (we put $\xi = 1$)

$$\int_0^\infty (P_{\mathcal{N}}(W) - P_{\mathcal{N}}^{(N)}(W))^2 \exp(-W)\, \mathrm{d}W = \sum_{n=N+1}^{\infty} (n!)^2 |c_n|^2 < \varepsilon, \tag{43}$$

where $P_{\mathcal{N}}^{(N)}$ denotes the function constructed as an Nth partial sum in (41), and ε is arbitrarily small.

Sometimes it is more suitable to use another decomposition of $P_{\mathcal{N}}(W)$ which has an explicit dependence on M:

$$P_{\mathcal{N}}(W) = W^{M-1} \sum_{j=0}^{\infty} c_j L_j^{M-1}(W), \tag{44}$$

where

$$c_j = \frac{j!}{(j+M-1)!} \sum_{s=0}^{j} \frac{(-1)^s}{(j-s)!\,(s+M-1)!} p(s). \tag{45}$$

Some examples demonstrating these methods will be given later.

If we want to determine the distribution $Q(W)$ from its moments $\langle W^k \rangle = \int_0^\infty Q(W)\, W^k\, \mathrm{d}W = q(k)$ it is sufficient to substitute $P_{\mathcal{N}}(W) \exp(-W) = Q(W)$ and $p(s)\, s! = q(s)$ into (44) and (45).

4. The Form of the Integrated Intensity Distribution

We shall now use some theorems about moments from statistics[40,41] to show clearly the conditions under which the integrated intensity distribution, given by (41), represents either an ordinary non-negative function or a generalized function whose support is composed of a finite number of points.

We define the moment sequence

$$M_n = \int_0^\infty W^n \, \mathrm{d}F(W), \qquad n = 0, 1, \ldots, \tag{46}$$

where $\mathrm{d}F(W) = F'(W)\,\mathrm{d}W = P_{\mathscr{N}}(W)\,\mathrm{d}W$. In the same way we can consider the sequence $\{p(n)\}$, denoting $p(n)n!$ by M_n and $\exp(-W)\,P_{\mathscr{N}}(W)$ by $F'(W)$. We now introduce the following system of quadratic forms[40]

$$Q_m = \sum_{i,k=0}^{m} M_{i+k}\, u_i\, u_k = \int_0^\infty \left(\sum_{i=0}^{m} W^i u_i \right)^2 \mathrm{d}F(W), \qquad m = 0, 1, \ldots, \tag{47}$$

where $\{u_i\}$ is an arbitrary non-trivial real vector. Thus the necessary and sufficient condition for $\mathrm{d}F(W)$ to be non-negative is $Q_m \geqslant 0$, for all m and an arbitrary non-trivial vector $\{u_i\}$. The condition $Q_m > 0$ is also necessary and sufficient for the existence of a non-decreasing function $F(W)$ $(0 \leqslant W < \infty)$ which fulfills (46) and has an infinite number of points at which the function increases.[40] Hence, if $Q_m > 0$ for all m and an arbitrary non-trivial vector $\{u_i\}$, then it is possible to construct the distribution $F'(W) \equiv P_{\mathscr{N}}(W) \geqslant 0$ from the given moment sequence $\{M_n\}_0^\infty$ in a unique way. Another case may arise if, for some m and some non-trivial vector $\{u_i\}$, the quadratic forms (47) equal zero. It can be shown[41] that in this case, a unique distribution $P_{\mathscr{N}}(W)$ exists, whose support is composed of a finite number of points. This is the minimum number of m for which $Q_m = 0$. On the other hand, if the function $P_{\mathscr{N}}(W) = F'(W)$ is concentrated at a finite number of points (say M) then $Q_m = 0$ for $m = M, M+1, \ldots$ and for a non-trivial vector $\{u_i\}$.

These results can be demonstrated by the following examples. Let us consider the system of determinants

$$\mathscr{D}_m = \begin{vmatrix} M_0 & M_1 & \ldots & M_m \\ M_1 & M_2 & \ldots & M_{m+1} \\ \ldots & \ldots & \ldots & \ldots \\ M_m & M_{m+1} & \ldots & M_{2m} \end{vmatrix}, \qquad m = 0, 1, \ldots \tag{48}$$

If $\mathscr{D}_m > 0$, then $Q_m > 0$. These conditions can easily be shown to be valid for thermal light for which $M_n = n!\,\langle n_T \rangle^n$, so that the function $P_{\mathscr{N}}(W)$ must exist as a unique ordinary function. (In this case $P_{\mathscr{N}}(W) = \langle n_T \rangle^{-1} \exp(-W/\langle n_T \rangle)$.) For the coherent field $M_n = \langle n_c \rangle^n$ and we see at once that

$\mathscr{D}_m = 0$ for $m \geqslant 1$ and $\mathscr{D}_0 = 1$. Thus, $Q_0 > 0$ and $Q_m = 0$ for $m \geqslant 1$. Using the above theorem, we find that the corresponding function $P_{\mathscr{N}}(W)$ is a generalized function having a one-point support. In fact, this distribution is equal to $\delta(W - \langle n_c \rangle)$.

Hence, if the moments M_n are known from experiment we should be able to decide, using the determinants (48) whether the distribution $P_{\mathscr{N}}(W)$ exists as an ordinary non-negative function or as a generalized function, whose support is composed of a finite number of points.

5. s-Ordering of Field Operators in Quantum Optics

Before demonstrating the methods developed above, we shall mention the problem of arbitrary ordering of field operators in quantum optics. This will enable us to give a number of examples in a very general form.

The problem of ordering field operators in quantum optics plays an increasing role in connection with generalized phase space distributions of optical fields[21,22,25,26,42], and also in laser theory.[43,44] Recently the problem of ordering field operators, and the correspondence between functions of c-numbers and functions of q-numbers has been systematically treated by Agarwal and Wolf.[16] (See also Lax.[45]) Their method was used for treating quantum dynamics problems in phase space, and for obtaining an expression for the time ordered product of Heisenberg operators, in terms of products ordered according to a prescribed rule (Wick's theorem can be deduced as a special case). A general treatment of this problem was also given by Cahill and Glauber.[17] They calculated quantum expectation values based on a kind of parametrization, which makes it possible to interpolate between normal and antinormal orderings, paying particular attention to the normal, symmetric (Weyl) and anti-normal orderings, and their differences and inter-relationships. These methods show when and how singularities appear in the corresponding weighting function, particularly in the weighting function of the diagonal representation of the density matrix in terms of the coherent states. An extension of this formulation to M-mode fields is also possible and fruitful in some cases, particularly for the superposition of coherent and thermal fields.[18,20]

This formulism is based on the decomposition of operators in terms of the so-called $\hat{T}$-operators which are defined by

$$\hat{T}(\beta, s) = \pi^{-1} \int \exp\left[\tfrac{1}{2}s|\alpha|^2 + \alpha(\hat{a}^\dagger - \beta^*) - \alpha^*(\hat{a} - \beta)\right] \mathrm{d}^2\alpha \qquad -1 \leqslant s \leqslant 1, \quad (49)$$

for which we have

$$\mathrm{Tr}\{\hat{T}(\alpha, s)\, \hat{T}(\beta, -s)\} = \pi\delta(\alpha - \beta). \quad (50)$$

The density operator $\hat{\rho}$ can be decomposed in terms of $\hat{T}$ in the form[16,17]

$$\hat{\rho} = \pi^{-1} \int \phi(\beta, -s)\, \hat{T}(\beta, s)\, \mathrm{d}^2\beta, \tag{51}$$

where

$$\phi(\beta, -s) = \mathrm{Tr}\{\hat{\rho}\, \hat{T}(\beta, -s)\}. \tag{52}$$

From (49) one can see that the $\hat{T}$-operator represents an s-ordered δ-function, and is equal to the δ-function for $s = 0$. The cases of normal, symmetric and antinormal orderings can be obtained for $s = 1, 0$ and -1. For example, for $s = -1$, $\hat{T}(\beta, -1) = |\beta\rangle\langle\beta|$ and (51) gives the diagonal representation in terms of coherent states.

Using such a formalism we can derive the following relation between s_1- and s_2-ordered generating functions[18]

$$\langle \exp(iy\hat{n})\rangle_{s_1} = \left(1 + \frac{s_1 - s_2}{2} iy\right)^{-M} \left\langle \exp\left[\frac{iy\hat{n}}{1 + [(s_1 - s_2)/2]\, iy}\right]\right\rangle_{s_2}. \tag{53}$$

The corresponding moments are

$$\langle \hat{n}^k\rangle_{s_1} = \frac{\mathrm{d}^k}{\mathrm{d}(iy)^k} \langle \exp(iy\hat{n})\rangle_{s_1}\Big|_{iy=0}$$

$$= \frac{(-1)^k k!}{(k + M - 1)!} \left(\frac{s_1 - s_2}{2}\right)^k \left\langle L_k^{M-1}\left(\frac{2\hat{n}}{s_1 - s_2}\right)\right\rangle_{s_2} \tag{54}$$

and the corresponding integrated intensity distributions are given by

$$P(W, s_1) = \frac{2}{s_2 - s_1} \int_0^\infty \left(\frac{W}{W'}\right)^{(M-1)/2} \exp\left[-\frac{2(W + W')}{s_2 - s_1}\right]$$

$$\times I_{M-1}\left(4\frac{(WW')^{\frac{1}{2}}}{s_2 - s_1}\right) P(W,' s_2)\, \mathrm{d}W' \qquad \text{for } s_2 > s_1, \tag{55}$$

where $I_{M-1}(x)$ is a modified Bessel function. It is also possible to derive the generalized photodetection equation[18]

$$p(n) = \frac{1}{(n + M - 1)!} \left(\frac{2}{1 + s}\right)^M \left(\frac{s - 1}{s + 1}\right)^n \int_0^\infty P(W, s)\, L_n{}^{M-1}$$

$$\times \left(\frac{4W}{1 - s^2}\right) \exp\left[-\frac{2W}{1 + s}\right] \mathrm{d}W. \tag{56}$$

Substituting $s = 1$, and using the asymptotic formula

$$L_n^{M-1}(x) \underset{x\to\infty}{\simeq} \frac{(n+M-1)!}{n!}(-x)^n, \tag{57}$$

we obtain, from (56) the standard photodetection equation

$$p(n) = \int_0^\infty P_{\mathcal{M}}(W)\frac{W^n}{n!}\exp(-W)\,\mathrm{d}W, \tag{58}$$

where $P_{\mathcal{M}}(W) \equiv P(W, 1)$.

6. Superposition of Coherent and Thermal Fields

We shall now give some physically interesting examples to demonstrate the methods developed above.

6.1. One-mode case

First we shall consider the superposition of coherent and thermal one-mode fields.

The Fock elements[46] for this case are

$$\rho(n,m) = \left(\frac{n!}{m!}\right)^{\frac{1}{2}} \exp\left[-\frac{|\beta|^2}{1+\langle n_T\rangle}\right]\frac{\langle n_T\rangle^n}{(1+\langle n_T\rangle)^{1+m}}\beta^{*m-n} \times \frac{1}{m!}L_n^{m-n}\left(-\frac{|\beta|^2}{\langle n_T\rangle(\langle n_T\rangle+1)}\right), \tag{59}$$

where $\langle n_T\rangle$ is the mean occupation number in the thermal field and β is the complex amplitude of the coherent field. We substitute these into (34) and obtain

$$c_{j\lambda} = \frac{\xi}{\pi(1+\langle n_T\rangle)^{j+\lambda+1}}\exp\left[-\frac{|\beta|^2}{1+\langle n_T\rangle}\right]\frac{j!}{[(j+\lambda)!]^3} \times [1+\langle n_T\rangle(1-\xi)]^j\beta^{*\lambda}L_j^{\lambda}\left(\frac{\xi|\beta|^2}{(1+\langle n_T\rangle)[1+\langle n_T\rangle(1-\xi)]}\right). \tag{60}$$

Then substituting this result into (32) and using the identities[30]

$$\sum_{n=0}^{\infty} \frac{n!}{[(n+\alpha)!]^3} L_n^{\alpha}(x) L_n^{\alpha}(y) z^n = \frac{(xyz)^{-\alpha/2}}{1-z} \times \exp\left[-z\frac{x+y}{1-z}\right] I_{\alpha}\left(2\frac{(xyz)^{\frac{1}{2}}}{1-z}\right), \tag{61}$$

$$I_{-\alpha}(x) = I_{\alpha}(x)$$

and

$$\sum_{n=-\infty}^{+\infty} I_n(x) \exp(in\phi) = \exp(x\cos\phi), \tag{62}$$

we obtain

$$\phi_{\mathcal{N}}(\alpha) = (\pi\langle n_T\rangle)^{-1} \exp\left[-\frac{|\alpha-\beta|^2}{\langle n_T\rangle}\right]. \tag{63}$$

This is the diagonal representation function for the superposition of coherent and thermal fields.

By using (61) and (62) we can easily verify that for the coefficients (60),

$$\sum_{j,\lambda} |c_{j\lambda}|^2 \frac{[(j+\lambda)!]^3}{j!} < \infty\,. \tag{64}$$

Note that the case of a coherent field can be treated as the limit $\langle n_T\rangle \to 0$ and that of a thermal field as the limit $\beta \to 0$.

6.2. MULTI-MODE CASE

The normal generating function for the superposition of coherent and thermal fields can be written in the form[47,19,33]

$$\left\langle \prod_{\lambda} \exp(ix\hat{n}_{\lambda}) \right\rangle_{\mathcal{N}} = \prod_{\lambda} (1 - ix\langle n_{T\lambda}\rangle)^{-1} \exp\left[\frac{ix\langle n_{c\lambda}\rangle}{1-ix\langle n_{T\lambda}\rangle}\right]. \tag{65}$$

Using the identities[30]

$$(1-ixB)^{-M} \exp\left[\frac{ixA}{1-ixB}\right] \equiv \sum_{n=0}^{\infty} \frac{(ixB)^n}{(n+M-1)!} L_n^{M-1}\left(-\frac{A}{B}\right) \tag{66a}$$

$$\equiv (1+B)^{-M} \exp\left[-\frac{A}{1+B}\right] \sum_{n=0}^{\infty} \frac{1}{(n+M-1)!} \times \left(1+\frac{1}{B}\right)^{-n} (1+ix)^n L_n^{M-1}\left(-\frac{A}{B(1+B)}\right), \tag{66b}$$

the following expressions can be obtained for the photon-counting distribution $p(n)$ and its factorial moments;

$$p(n) = \frac{1}{n!}\frac{\mathrm{d}^n}{\mathrm{d}(ix)^n}\left\langle \prod_\lambda \exp(ix\hat{n}_\lambda)\right\rangle_{\mathscr{N}}\Big|_{ix=-1}$$

$$= \prod_\lambda (1 + \langle n_{T\lambda}\rangle)^{-1} \exp\left[-\frac{\langle n_{c\lambda}\rangle}{1 + \langle n_{T\lambda}\rangle}\right]$$

$$\times \sum_{\substack{\lambda \\ \sum n_\lambda = n}} \prod_\lambda \frac{1}{n_\lambda!}\left(1 + \frac{1}{\langle n_{T\lambda}\rangle}\right)^{-n_\lambda} L_{n_\lambda}^0\left(-\frac{\langle n_{c\lambda}\rangle}{\langle n_{T\lambda}\rangle(1 + \langle n_{T\lambda}\rangle)}\right) \tag{67}$$

and

$$\langle W^k\rangle_{\mathscr{N}} = \frac{\mathrm{d}^k}{\mathrm{d}(ix)^k}\left\langle \prod_\lambda \exp(ix\hat{n}_\lambda)\right\rangle_{\mathscr{N}}\Big|_{ix=0}$$

$$= k! \sum_{\substack{\lambda \\ \sum m_\lambda = k}} \prod_\lambda \frac{1}{m_\lambda!}\langle n_{T\lambda}\rangle^{m_\lambda} L_{m_\lambda}^0\left(-\frac{\langle n_{c\lambda}\rangle}{\langle n_{T\lambda}\rangle}\right). \tag{68}$$

The corresponding integrated intensity distribution can in principle be obtained from (44);

$$P_{\mathscr{N}}(W) = W^{M-1}\sum_{j=0}^{\infty}\frac{j!\,L_j^{M-1}(W)}{(j + M - 1)!}\sum_{s=0}^{j}\frac{(-1)^s}{(j-s)!(s + M - 1)!}p(s). \tag{69}$$

From these general results one can obtain, for example, the formulae for the detection of a thermal field using a heterodyne technique. In this method, the thermal field is superimposed on a known coherent component. Jakeman and Pike,[8] who proposed this method, pointed out that additional spectral information can be obtained, i.e. the central frequency of the thermal field can also be determined by this method. In general, the central frequency of the thermal field and the frequency of the coherent field are arbitrary and different from one another. We must therefore consider the superposition of coherent and thermal fields of different mean frequencies. The results can be obtained from equations (67), (68) and (69), taking $\lambda = 1, 2, \ldots, M$ for M modes with $\langle n_{T\lambda}\rangle = \langle n_T\rangle/M$, and $\lambda = M + 1, \ldots, 2M$ for M modes with $\langle n_{T\lambda}\rangle = 0$.

We may write

$$\sum_{\substack{\lambda \\ \sum n_\lambda = n}}^{2M} = \sum_{j=0}^{n}\sum_{\substack{\lambda \\ \sum n_\lambda = j}}^{M}\sum_{\substack{\lambda = M+1 \\ \sum n_\lambda = n-j}}^{2M} \tag{70}$$

Using the polynomial theorem and an asymptotic expression for the L_m^0-polynomials, together with the following identity for the Laguerre polynomials[48]

$$\sum_{\substack{M \\ \sum_\lambda n_\lambda = j}} \prod_\lambda^M \frac{1}{(m_\lambda + \alpha_\lambda)!} L_{m_\lambda}^{\alpha_\lambda}(x_\lambda) = \frac{1}{\left(j + \sum_\lambda^M \alpha_\lambda + M - 1\right)!} L_j^{\sum_\lambda^M \alpha_\lambda + M - 1}\left(\sum_\lambda^M x_\lambda\right), \tag{71}$$

we obtain[20]

$$p(n) = \left(1 + \frac{\langle n_T\rangle}{M}\right)^{-M} \exp\left[-\frac{\langle n_c\rangle[M + \langle n_T\rangle(1-\omega^2)]}{M + \langle n_T\rangle}\right] \times \sum_{j=0}^{n} \frac{1}{(n-j)!(j+M-1)!}[\langle n_c\rangle(1-\omega^2)]^{n-j} \times \left(1 + \frac{M}{\langle n_T\rangle}\right)^{-j} L_j^{M-1}\left(-\frac{\langle n_c\rangle \omega^2 M^2}{\langle n_T\rangle(\langle n_T\rangle + M)}\right) \tag{72}$$

and

$$\langle W^k\rangle_{\mathcal{N}} = \sum_{j=0}^{k} \frac{k!}{(k-j)!(j+M-1)!}[\langle n_c\rangle(1-\omega^2)]^{k-j} \times \left(\frac{\langle n_T\rangle}{M}\right)^j L_j^{M-1}\left(-\frac{\langle n_c\rangle \omega^2 M}{\langle n_T\rangle}\right), \tag{73}$$

where $\sum_\lambda^M \langle n_{c\lambda}\rangle = \langle n_c\rangle \omega^2$ and $\sum_{\lambda=M+1}^{2M} \langle n_{c\lambda}\rangle = \langle n_c\rangle(1-\omega^2)$.

In these equations the parameter $\omega = [2\sin(\Omega/2)]/\Omega$, where Ω is equal to the difference between the mean frequency of the thermal light and the frequency of the coherent light multiplied by the time interval T of observation.

For $P_{\mathcal{N}}(W)$ we obtain

$$P_{\mathcal{N}}(W) = \begin{cases} \dfrac{M}{\langle n_T\rangle}\left(\dfrac{W - \langle n_c\rangle(1-\omega^2)}{\langle n_c\rangle \omega^2}\right)^{(M-1)/2} \times \exp\left[-\dfrac{W + \langle n_c\rangle(2\omega^2 - 1)}{\langle n_T\rangle} M\right] \\ \qquad \times I_{M-1}\left(2\omega M \dfrac{[\langle n_c\rangle(W - \langle n_c\rangle(1-\omega^2))]^{\frac{1}{2}}}{\langle n_T\rangle}\right) \\ \qquad \text{for } W > \langle n_c\rangle(1-\omega^2) \\ 0 \text{ for } W < \langle n_c\rangle(1-\omega^2). \end{cases} \tag{74}$$

The s-ordering formalism mentioned above gives us the more general formulae

$$\langle W^k\rangle_s = \frac{k!}{(k+M-1)!}\left(\frac{1-s}{2}\right)^k \sum_{j=0}^{k} \frac{1}{(j+M-1)!}\left(\frac{2\langle n_T\rangle}{(1-s)M}\right)^j \times L_j^{M-1}\left(-\frac{\langle n_c\rangle \omega^2 M}{\langle n_T\rangle}\right) L_{k-j}^{j+M-1}\left(-\frac{2\langle n_c\rangle(1-\omega^2)}{1-s}\right) \tag{75}$$

and

$$P(W,s) = \left(\frac{W}{\langle n_c\rangle(1-\omega^2)}\right)^{(M-1)/2}\left(\frac{\langle n_T\rangle}{M}+\frac{1-s}{2}\right)^{-M}\left(\frac{1-s}{2}\right)^{M-1} \times \exp\left[-\frac{2W}{1-s}-\frac{\langle n_c\rangle}{\langle n_T\rangle/M+(1-s)/2} - \frac{2\langle n_c\rangle\langle n_T\rangle(1-\omega^2)}{M(1-s)[\langle n_T\rangle/M+(1-s)/2]}\right] \times \sum_{l=0}^{\infty}\frac{1}{(l+M-1)!}\left[\left(\frac{W}{\langle n_c\rangle(1-\omega^2)}\right)^{\frac{1}{2}}\frac{\langle n_T\rangle}{M} \times \left(\frac{\langle n_T\rangle}{M}+\frac{1-s}{2}\right)^{-1}\right]^l I_{l+M-1}\left(4\frac{(W\langle n_c\rangle(1-\omega^2))^{\frac{1}{2}}}{1-s}\right) \times L_l^{M-1}\left(-\frac{(1-s)\omega^2 M\langle n_c\rangle}{2\langle n_T\rangle[\langle n_T\rangle/M+(1-s)/2]}\right). \tag{76}$$

A number of special cases can be obtained putting $\omega = 1$, 0 $s = 1, 0, -1$ (normal, symmetric and antinormal orderings), $M = 1$, etc. For example, for the case where the mean frequencies of the thermal and coherent fields are identical, i.e. $\omega = 1$, we have[18]

$$\langle W^k\rangle_s = \frac{k!}{(k+M-1)!}\left(\frac{\langle n_T\rangle}{M}+\frac{1-s}{2}\right)^k L_k^{M-1}\left(-\frac{\langle n_c\rangle}{\langle n_T\rangle/M+(1-s)/2}\right) \tag{77}$$

and

$$P(W,s) = \left(\frac{\langle n_T\rangle}{M}+\frac{1-s}{2}\right)^{-1}\left(\frac{W}{\langle n_c\rangle}\right)^{(M-1)/2}\exp\left(-\frac{(W+\langle n_c\rangle)}{\langle n_T\rangle/M+(1-s)/2}\right) \times I_{M-1}\left(2\frac{(\langle n_c\rangle W)^{\frac{1}{2}}}{\langle n_T\rangle/M+(1-s)/2}\right). \tag{78}$$

The case $s = -1$ would be useful for determining the statistical properties of light using "quantum counters".[23]

More general results can be gained from the s-ordered generating function

$$\langle \exp ix\hat{n} \rangle_s = \prod_\lambda \left[1 - ix\left(\langle n_{T\lambda}\rangle + \frac{1-s}{2}\right)\right]^{-1} \times \exp\left[\frac{ix\langle n_{c\lambda}\rangle}{1 - ix(\langle n_{T\lambda}\rangle + (1-s)/2}\right]. \quad (79)$$

We obtain

$$\langle W^k \rangle_s = k! \sum_{\sum_\lambda n_\lambda = k} \prod_\lambda \frac{1}{n_\lambda!}\left[\langle n_{T\lambda}\rangle + \frac{1-s}{2}\right]^{n_\lambda} L_{n_\lambda}^0\left(-\frac{\langle n_{c\lambda}\rangle}{\langle n_{T\lambda}\rangle + (1-s)/2}\right). \quad (80)$$

The general integrated intensity distribution can be obtained from $\langle W^k \rangle_s$;

$$P(W, s) = W^{M-1} \exp(-W) \sum_{n=0}^{\infty} c_n L_n^{M-1}(W), \quad (81)$$

where

$$c_n = \frac{n!}{(n + M - 1)!} \sum_{j=0}^{n} \frac{(-1)^j}{j!(n-j)!(j+M-1)!} \langle W^j \rangle_s, \quad (82)$$

with the condition that

$$\int_0^\infty P^2(W, s)\, W^{1-M} \exp(W)\, dW = \sum_{n=0}^{\infty} \frac{[(n + M - 1)!]^3}{n!} |c_n|^2 < \infty. \quad (83)$$

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Particle Beam Fluctuations in Quantum Mechanics

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Photon beam fluctuations have been studied by many authors. Information about the statistical properties defining a photon beam may be obtained by coincidence and counting experiments and the properties are found to vary from those of Gaussian light to those of laser light. We know how to explain these results for bosons in quantum optics by using a theory which enables us to describe both the field and the detector acting on the field. The results are well known and we shall now ask what statistical properties we might expect to observe if a counting or a coincidence experiment is performed on a beam of fermions. Can the statistical properties of fermion beams vary over a range comparable with that of photons from Gaussian or other sources? This question is very broad and we shall not provide a complete answer. The point we wish to stress here is the connection between the various statistical properties that may be detected in a system of particles and the quantum nature of the particles.

We shall define the statistical properties of a beam of particles in terms of its measured coincidence probabilities. The coincidence probability $P_p(a_1, t_1, \ldots, a_p, t_p)$ is the probability that any particle will be detected at point a_1 at time $t_1, \ldots$, any other at point a_p at time t_p, where p is any positive integer. This choice of the coincidence probability is determined by two considerations. The first is that the set of the coincidence probabilities should completely define the statistical properties of the beam (including their time dependence), and the second is that they may be measured directly.

In order to describe the system of particles and their detection, and to calculate these probabilities, we shall use the wave packet formalism. This formalism enables us to describe and compare the behaviour of boson and fermion beams in coincidence experiments. The formalism was first used by Goldberger and Watson[1] to calculate the second order coincidence probability for a particle beam, but they used a normalization which is not strictly valid.

1. The Wave Packet Formalism

Consider a system of indistinguishable and non-interacting particles. The number of particles, $N(\omega)$, is a random variable. A stochastic wave packet

$\Phi_i(r, t, \omega_i')$ is associated with every particle of the beam located at r at time t. The choice of a value of the random variable ω_i' corresponds to the choice of Φ_i among a given set of functions (depending on the beam). All the Φ_i are identical stochastic functions. We consider the set of particles as resulting from the superposition of states (ω, ω'), where the number of particles and the wave packets are given. Each of these states is described by a given wave function, ψ. Since the particles do not interact, we can calculate ψ from the functions Φ_i. The beam is described by the stochastic function $\psi[N(\omega), r_1, \ldots, r_{N(\omega)}, t, \omega']$ depending on the random variable ω and on the set of random variables $\omega' = \{\omega_i'\}$. All these variables may, or may not, be independent.

2. Ideal Detectors

Consider an ideal two-level atom used as a photon detector (see chapter by Glauber in this volume). This detector is assumed to act in a volume, and with a response time very much smaller than the volumes and times characterizing the field. Such a detector placed in the field tells us about the presence, or otherwise, of a particle at point a, at time t, i.e. it projects the system at time t from the state ψ, which it may occupy before the measurement, into a subspace $\mathscr{E}(a, t)$ of the space describing the states of the system. $\mathscr{E}(a, t)$ contains all the states of the system for which we know that a particle is at point a at time t. The probability for such a projection to occur is given by the first order coincidence probability. We can make a measurement by using p detectors located at points $a_1, \ldots, a_p$ at times $t_1, \ldots, t_p$; each detector acts as a projection operator in a sub-space $\mathscr{E}(a_i, t_i)$. The probability for detection of a particle by every detector is the pth order coincidence function $P_p[a_1, t_1, \ldots, a_p, t_p]$. For fermions, ideal detectors based on secondary emission, for instance, may be devised. For bosons and fermions, then, it is theoretically possible to locate a particle in space and time and so to measure coincidence probabilities.

3. General Formulation of Coincidence Probabilities

3.1. Coincidence probabilities at a given time

Consider a system consisting of one particle described by a stochastic wave function $\Phi(r, t, \omega')$. The ensemble average $\langle|\Phi(a, t)|^2\rangle$† gives the probability of detecting the particle at point a, at time t, where Φ is normalized on a certain volume V where the particle is known to be.

If there are $N(\omega)$ particles inside V, the probability of finding a particle at a_1, any other at a_2, the Nth at a_N, at time t, while the system is in state (ω, ω') is

† The symbol $\langle \quad \rangle$ indicates an ensemble average.

$P_{N,(\omega,\omega')}(a_1, \ldots, a_N, t)$ and is such that

$$\int_{r_1 \leqslant r_2 \leqslant \ldots \leqslant r_n} \int \ldots \int P_{N,(\omega,\omega')}[r_1, \ldots, r_N, t]\, dr_1 \ldots dr_N = 1, \tag{1}$$

where $r_1, \ldots, r_N(V)$. In performing this integration, we are considering all the possible different N coincidence measurements that we can make on this set of particles. We know that by these measurements, we are sure to detect the N particles of the system, this justifies eqn (1).

Since the normalization condition on ψ may be written

$$\int_V |\psi|^2\, dr_1 \ldots dr_N = 1, \tag{2}$$

it is easy to show that

$$P_{N,(\omega,\omega')}[a_1, \ldots, a_N, t] = N(\omega)!\, |\psi[N(\omega), a_1, \ldots a_N, t, \omega']|^2. \tag{3}$$

However, we are interested in more general coincidence probabilities, viz. the probabilities of detecting p particles, without knowing the total number of particles. To determine these probabilities, we just calculate the conditional probabilities of detecting p particles at $a_1, \ldots, a_p$ at time t, knowing that there are $N(\omega)$ particles in the system. The probability we are concerned with is an *a priori* probability obtained from the conditional probability by taking the mean value over ω and ω'. We obtain

$$P_p(a_1, \ldots, a_p, t) = \left\langle \int_V \int_V |\psi|^2 \sum_{\beta_1 \neq \beta_2 \neq \ldots \neq \beta_p} \prod_{i=1}^{p} \delta(a_i - r_{\beta_i})\, dr_1 \ldots dr_N \right\rangle \tag{4}$$

$(\beta_1, \ldots, \beta_p)$ are permutations of p elements taken in N elements.

3.2. COINCIDENCE PROBABILITIES AT DIFFERENT TIMES

Using the Dirac formalism, the wave function ψ is written as a vector $|\psi\rangle$ of the state space. We can describe the action of p detectors acting at times $t_1, < t_2, < \ldots, < t_p$ by an operator. This operator is the product of the projection operators on $\mathscr{E}(a_i, t_i)$, $\mathscr{P}(a_i, t_i)$. The probability may be written

$$\langle (\psi | \mathscr{P}(a_1, t_1) \ldots \mathscr{P}(a_p, t_p) \ldots \mathscr{P}(a_1, t_1) | \psi) \rangle.$$

In the general case considered so far, this expression is quite complicated but, in the stationary incoherent case described below, it can be shown that such a probability may be obtained formally from the coincidence probability, $P_p(a_1, a_2, \ldots, a_p, t)$. In fact $P_p(a_1, \ldots, a_p, t)$ may be described completely in terms of the function $\Phi_i(a_i, t, \omega_i')$ and $P_p(a_1, t_1; \ldots; a_p\, t_p)$ may be written in exactly the same way only by changing $\Phi_i(a_i, t, \omega_i')$ into $\Phi_i(a_i, t_i, \omega_i')$. So, in

the following we shall only consider eqn (4). Any other coincidence probability can be easily obtained from the results we get.

4. Coincidence Probability in the Stationary Incoherent Case

To calculate $P_p(a_1, t_1, \ldots, a_p, t_p)$ given by eqn (4) we have first to expand ψ in terms of Φ. Since ψ is normalized (eqn (2)), it is written as

$$\psi = \Sigma P_N{}^K(\pm 1)^K \prod_{i=1}^{N} \Phi_{\alpha_i}(r_i, t, \omega') \times \left\{ \left[\int_V \ldots \int_V \Sigma P_N{}^{K'}(\pm 1)^{K'} \prod_{i=1}^{N} \Phi_{\beta_i}(r_i, t, \omega') \times \Sigma P_N{}^{K''}(\pm 1)^{K''} \prod_{i=1}^{N} \Phi_{\gamma_i}{}^*(r_i, t, \omega')\, dr_1 \ldots dr_N \right]^{-\frac{1}{2}} \right\}^{-1} \tag{5}$$

$P_N{}^K$ is the sum of all the permutations of order k, of $\alpha_1, \ldots, \alpha_N$.

If the functions Φ_i were orthogonal, the denominator of eqn (5) would reduce to $\sqrt{N!}$. Goldberger and Watson assumed that this was the case, but it can be shown that this is valid only in certain cases. We cannot assume, *a priori*, that the functions Φ_i are orthogonal for all types of beam. On the contrary, the Φ_i, being stochastic identical functions, it can be shown that it is impossible for them to be orthogonal for every set of $\langle \omega_i' \rangle = \omega'$. Therefore, P_p must be written in a general way thus,

$$P_p = \left\langle \left[\int_V \ldots \int_V \prod_{i=1}^{N} \Phi_i(r_i, t, \omega') \Sigma P_N{}^K(\pm 1)^K \Phi_{\alpha_i}{}^*(r_i, t, \omega') \times \sum_{\beta_1 \neq \ldots \neq \beta_p} \prod_{j=1}^{p} \delta(a_j - r_{\beta_j})\, dr_1 \ldots dr_N \right] \times \left[\int_V \ldots \int_V \prod_{i=1}^{N} \Phi_i(r_i, t, \omega') \Sigma P_N{}^{K'}(\pm 1)^{K'} \times \Phi_{\alpha_i}{}^*(r_i, t, \omega')\, dr_1 \ldots dr_N \right]^{-1} \right\rangle. \tag{6}$$

This formal result is too general to tell us much about the properties of P_p.

For the case of a completely incoherent stationary beam, it can be shown that the denominator, D, of eqn (6) cancels with factors in the numerator of (6). The probabilities P_p are then rather simple and several remarks can be made about them. We define a completely incoherent beam as follows:

a. the Φ_i are identical and stochastically independent;

b. the stochastic laws of $N(\omega)$ and of the Φ_i are independent;

c. the beam is stationary in time and space (in particular we consider such an extension of the beam that the spreading of the wave packets may be neglected.

To study this stationary incoherent system, we shall consider the two different cases which cover the few experimental situations.

1. The number of particles, δ, passing through the coherence area during the coherence time τ is very much less than 1. This condition is satisfied by monochromatic natural light ($\delta \approx 10^{-3}$) and by the most monoenergetic and powerful electron beams we can produce ($\delta \approx 10^{-4}$–10^{-5}). In this case, $D \approx 1$.

2. When δ is not small compared to 1, the total mean number of particles, $\langle N \rangle$, is quite large. By using the law for large numbers, we can show that the factors appearing in the numerator cancel with D, in (6). Then $D \gg 1$, and the calculation of Goldberger and Watson is not valid.

In both cases (1 and 2), we obtain the same expression for P_p,

$$P_p(a_1, t_1, \ldots, a_p, t_p) = \langle N \rangle^p \left\langle \prod_{i=1}^{p} \Phi_i(a_i, t_i, \omega_i') \times \sum_{K=0}^{N-1} P_p{}^K (\pm 1)^K \prod_{i=1}^{p} \Phi_{\alpha_i}{}^* (a_i, t_i, \omega_i') \right\rangle . \tag{7}$$

If we set

$$\eta_i(a, t, \omega_i') = \Phi_i(a, t, \omega_i') \langle |\Phi_i|^2 \rangle^{-\frac{1}{2}},$$

we can rewrite P_p in the form

$$P_p = \rho^p \left\langle \prod_{i=1}^{p} \eta_i(a_i, t_i, \omega_i') \, \Sigma P_p{}^K (\pm 1)^K \prod_{i=1}^{p} \eta_{\alpha_i}{}^* (a_i, t_i, \omega_i') \right\rangle,$$

where ρ is the mean density of particles in the system.

5. Discussion

a. We first note that in the range of the possible beams considered (non-interacting particles), the statistical properties do not depend on the strength of the beam.

b. Putting $p = 2$ in eqn (7), we get, (see Fig. 1 for $|\gamma_{12}| = \exp(-2|t_1 - t_1|/\tau)$)

$$P_2(a_1, t_1, a_2, t_2) = \rho^2 [1 \pm |\gamma_{12}|^2] \tag{8}$$

where

$$\gamma_{12} = \langle \eta(a_1, t_1)\, \eta^*(a_2, t_2) \rangle = \gamma_{21}{}^*.$$

Equation (8) gives the well-known bunching effect for bosons (+ sign). For fermions (− sign), it shows an "antibunching effect". In the latter case, the conditional probability of detecting a particle at a given point at a given time when we have already detected another particle at a neighbouring point and time (i.e. within the coherence volume and coherence time) is smaller than the *a priori* probability of detecting a particle within the coherence volume during the coherence time. This second order effect could have been predicted easily from our knowledge of fermions. The interest in our result lies in its application to higher order coincidence probabilities.

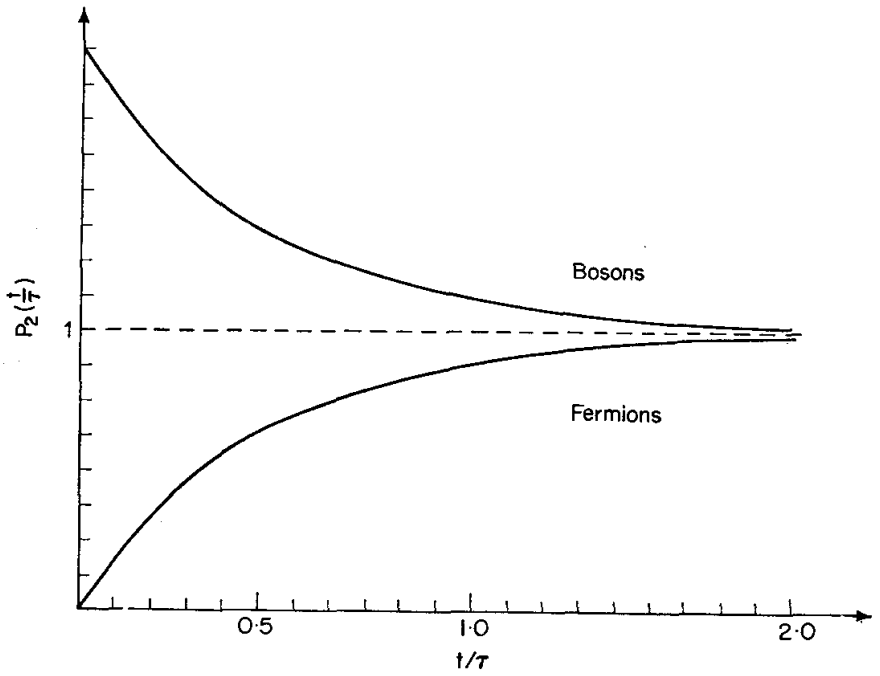


FIG. 1

c. Equation (7) shows that both the bunching effect and the antibunching effect occurring in an incoherent beam of particles come from the terms corresponding to non-zero order permutation. This means that, if the particles were distinguishable, the P_p would merely define a Poisson process. But, because of their indistinguishability, the particles which are assumed to be stochastically independent when emitted, can only be detected as bunched or antibunched.

d. All the well-known results for bosons are obtainable from eqn (7) when the positive sign is taken. The P_p obtained are the coincidence probabilities of

a compound Poisson process. They are equal to the moments of a stochastic real positive function $\mathscr{T}(r, t)$ given by

$$\mathscr{T}(r, t) = E(r, t)\, E^*(r, t), \tag{9}$$

where $E(r, t)$ is a Gaussian stochastic function whose correlation function is given by

$$\langle E(a_1, t,)\, E^*(a_2, t_2)\rangle = \rho\gamma_{12}.$$

It can be considered as the field associated with the particles.

e. Fermions have several interesting features.

1. Consider the one-dimensional stochastic process which is a function of time only, obtained by making all the coincidence measurements at the same point in space. Assuming that the correlation function

$$\gamma_{12} = \langle \eta(a, t_1)\, \eta^*(a, t_2)\rangle$$

is such that

$$\gamma_{13} = \gamma_{12}\gamma_{23} \quad \text{for} \quad t_1 \leqslant t_2 \leqslant t_3, \tag{10}$$

the process is a *renewal process*. It means that the waiting time between two detected particles does not depend on the particles detected previously. This result depends on the assumption that the particles do not interact. The probability for the waiting time to be equal to t, if $|\gamma_{12}| = \exp(-2|t_1 - t_2|/\tau)$ is given by (see Fig. 2)

$$p\frac{t}{\tau} = \frac{2\rho \exp(-t/\tau) \sinh[(t/\tau)(1-2\rho\tau)^{\frac{1}{2}}]}{(1-2\rho\tau)^{\frac{1}{2}}} \tag{11}$$

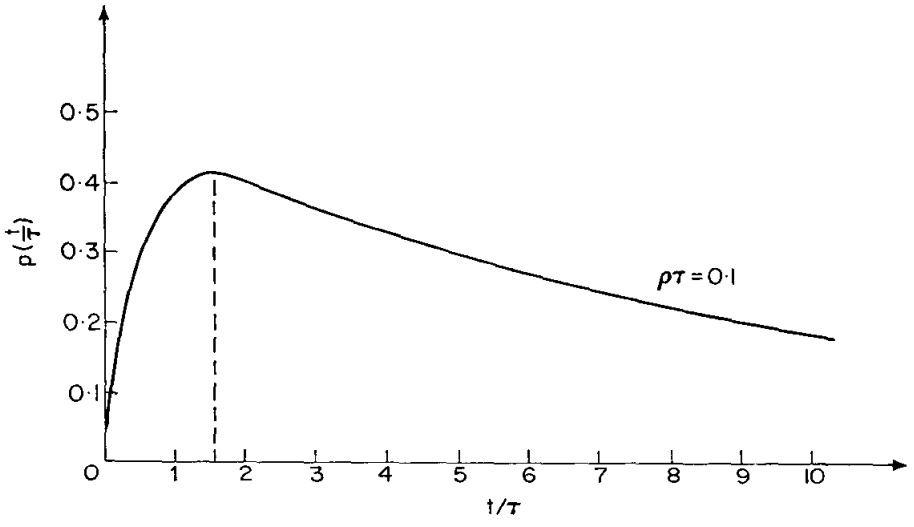


FIG 2.

From this it follows that $2\rho\tau \leqslant 1$; this is simply an expression of the Pauli exclusion principle.

2. We may also study the statistical properties of the number of particles that can be detected at a given point during a time, T. The probability $P_n(T)$ of detecting n particles during time T can be obtained from the set of functions P_p. We see that for $n \geqslant 2$ and $T \ll \tau$, we have $P_n(T) \leqslant 0$. Again, this is the Pauli exclusion principle. For $T > \tau$ (i.e. $T \gtrsim 3\tau$ or 4τ) we shall consider three stochastic processes, the mean intensities of which are the same. We shall assume that one is a Poisson process, the second is a Gaussian Poisson compound process, and the third is the detected process of a stationary incoherent beam of fermions defined by the same second order correlation function γ_{12} as the compound Poisson process. Then we have

$$P_{nF}(T) = 2P_{nP}(T) - P_{nB}(T), \tag{12}$$

where P_{nB} refers to the Poisson compound process, P_{nP} to the Poisson process, and P_{nF} to the fermion process. This symmetry between fermions and bosons is only an approximate one, that is allowable for $T \gtrsim 3\tau$ or 4τ (then the relative error is $\sim 10^{-3}$). The symmetry is completely true only for the second order statistical properties of the beam, viz. the second order coincidence probability, or the variance of the number n detected during T, for instance (see Figs. 3 and 4).

3. We shall now consider the possibility of performing experiments to observe these statistical properties of a beam of fermions. Electrons appear to offer the most favourable conditions, with the power and monochromaticity of the sources now available. But these sources, if they give a coherence area large enough to allow good measurements, give a coherence time of the order of 10^{-13} sec. The fastest electron detectors available are generally 3 or 4 orders of magnitude slower than this, so they would completely smooth the effects we should like to observe. This situation is similar to that which confronts us when we wish to perform correlation or coincidence experiments on white light.

The fermion problem we have considered seems to be beyond the reach of experimental confirmation at present.

6. Conclusions

The formalism we have used has shown in a very simple way how the bunching or antibunching effect comes from the *detection* of indistinguishable particles. This effect is very important when the particles are stochastically independent. It can be cancelled out, for bosons, if certain correlations are assumed between the particles (for example, as in the laser).

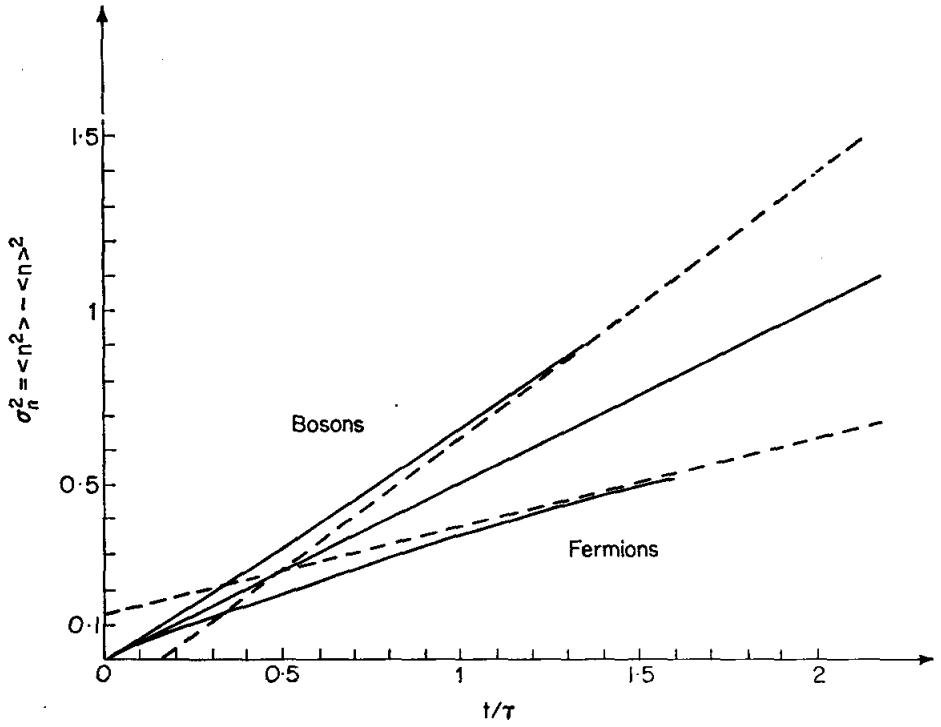

$\sigma_n^2 = \langle n^2 \rangle - \langle n \rangle^2$
1·5
1
0·5
0·1
0
0·5
1
1·5
2
t/τ
Bosons
Fermions

FIG. 3

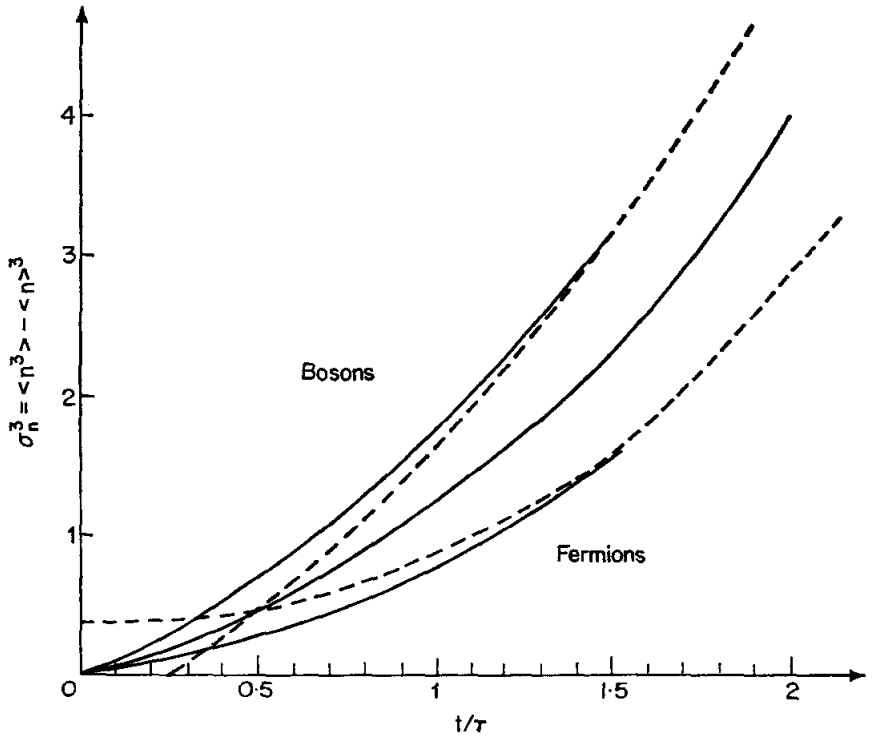

$\sigma_n^3 = \langle n^3 \rangle - \langle n \rangle^3$
4
3
2
1
0
0·5
1
1·5
2
t/τ
Bosons
Fermions

FIG. 4

We can also analyse completely the incoherent stationary fermion-beam and stress several interesting characteristic features of it. It can be shown that this incoherent stationary case corresponds to thermal equilibrium.

Now, it would be interesting, from a theoretical point of view, to have a broader view of the question, that is, to analyse how the statistical properties of a fermion-beam vary according to the different sources that could be considered. In particular, the case of fermion-beams not in thermal equilibrium should also be studied.

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